PB81-106239

Technical Support of Standards for High-Level Radioactive Waste Management. Volume B Engineering Controls

Arthur D. Little, Inc.
Cambridge, MA

Prepared for

Office of Radiation Programs
Washington, DC

1979

U.S. Department of Commerce
National Technical Information Service
NTIS

TECHNICAL REPORT DATA

(Please read Instructions on the reverse before completing)

1. REPORT NO.
EPA 520/4-79-007B

2.

3. RECIPIENT'S ACCESSION NO.
PB81 106239

4. TITLE AND SUBTITLE
Technical Support of Standards for High-level Radioactive Waste Management, Volume B, Engineering Controls

5. REPORT DATE
1979

6. PERFORMING ORGANIZATION CODE

7. AUTHOR(S)

8. PERFORMING ORGANIZATION REPORT NO.

9. PERFORMING ORGANIZATION NAME AND ADDRESS
Arthur D. Little, Inc.
Cambridge, Massachusetts 02140

10. PROGRAM ELEMENT NO.

11. CONTRACT/GRANT NO.
68-01-4470

12. SPONSORING AGENCY NAME AND ADDRESS
Office of Radiation Programs
Environmental Protection Agency
Washington, D.C. 20460

13. TYPE OF REPORT AND PERIOD COVERED
March - August 1977

14. SPONSORING AGENCY CODE
ANR-460

15. SUPPLEMENTARY NOTES

16. ABSTRACT

This report is the result of work performed under the second part (Task B) of a four-part contract to gather technical information to evaluate environmental acceptability of various options for disposing high-level wastes. The other tasks are: A-Source Term Characterization; C-Assessment of Migration Pathways; D-Assessment of Accidental Pathways.

The scope of work for Task B was divided into 2 major steps: analysis of technology for engineering control of high-level wastes, and projections of costs for various alternative disposal technologies. Primary emphasis has been placed in non-reprocessed spent fuel. Brief consideration has been given to lower-level transuranic contaminated wastes which arise primarily from plutonium fuel fabrication and reprocessing plants.

The objective of this limited review is to provide some insight into the state of the art for applying engineering controls to such wastes, including, where necessary, direct burial with high-level wastes in a geologic disposal facility.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
high-level wastes radioactive waste disposal radioactive waste management engineering controls for high-level wst dis waste disposal technologies		

18. DISTRIBUTION STATEMENT

19. SECURITY CLASS *(This Report)*

20. SECURITY CLASS *(This page)*

21. NO. OF PAGES

22. PRICE

EPA Form 2220-1 (Rev. 4-77) PREVIOUS EDITION IS OBSOLETE

United States
Environmental Protection
Agency

Office of
Radiation Programs
Washington DC 20460

EPA 520/4-79-007B

PB81-106239

Radiation

EPA

Technical Support of Standards for High-Level Radioactive Waste Management

Volume B

Engineering Controls

REPRODUCED BY
NATIONAL TECHNICAL
INFORMATION SERVICE
U. S. DEPARTMENT OF COMMERCE
SPRINGFIELD, VA. 22161

TECHNICAL SUPPORT OF STANDARDS FOR HIGH-LEVEL RADIOACTIVE WASTE MANAGEMENT

TASK B REPORT

EPA Contract No. 68-01-4470

Prepared by

Arthur D. Little, Inc.
Cambridge, Massachusetts 02140
March-August 1977

DISCLAIMER

This report was prepared as an account of work sponsored by the Environmental Protection Agency of the United States Government under Contract No. 68-01-4470. Neither the United States nor the United States Environmental Protection Agency makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights.

ACKNOWLEDGEMENTS

Many individuals contributed to the work done under the direction of Arthur D. Little, Inc., for the U.S. Environmental Protection Agency under Contract No. 68-01-4470. John L. Russell and Daniel Egan of the Office of Radiation Programs at EPA served as constant guides in the process of our work. Dr. Bruce S. Old, James I. Stevens, and David I. Hellstrom of Arthur D. Little, Inc., were Program Director, Program Manager, and Assistant Program Manager, respectively, of the overall project. Key individuals involved in each of the reports prepared under the four tasks were:

TASK A	Donald Korn Arthur D. Little, Inc. Task Director
	Robert McWhorter, Michael Raudenbush, and Lester Goldstein S.M. Stoller Corp.
TASK B	Edwin L. Field Arthur D. Little, Inc. Task Director
	Robert McWhorter and Michael Raudenbush S.M. Stoller Corp.
TASK C	P.J. O'Brien Arthur D. Little, Inc. Task Director
	Dr. Ronald B. Lantz Intera Environmental Consultants, Inc.
	Dr. John Gormley D'Appolonia Consulting Engineers, Inc.
TASK D	Donald S. Allan Arthur D. Little, Inc. Task Director
	Ajit Bhattacharyya and Charles R. Hadlock Arthur D. Little, Inc.

FOREWORD

A major Federal effort is underway to develop methods for disposal of high-level radioactive waste in deep geologic repositories. An important element of this program is the development and promulgation by the U.S. Environmental Protection Agency (EPA) of environmental standards for the management of these wastes.

In anticipation of its efforts to develop these standards, EPA recognized that it would be necessary to estimate the expected and potential environmental impacts from potential geologic repositories using modeling techniques based upon as thorough an understanding as possible of the uncertainties involved in the quantities and characteristics of the wastes to be managed, the effectiveness of engineering controls, and the potential migration and accidental pathways that might result in radioactive materials entering the biosphere. Consequently, in March 1977, the EPA contracted with Arthur D. Little, Inc.,for a study to provide technical support for its development of environmental regulations for high-level radioactive wastes. This study was divided into the following four tasks:

Task A - Source Term Characterization/Definition

B - Effectiveness of Engineering Controls

C - Assessment of Migration Pathways

D - Assessment of Accidental Pathways

The information presented in the reports on these tasks was developed principally during the period March 1977 to February 1978. In the case of this report, Task B, the information contained in it was prepared during the period March-August 1977. There are many national and international programs underway to develop additional data, especially in the fields of waste forms, knowledge of geology and geohydrology, and risk assessment. The information presented in these reports has been developed on conceptual bases and is not intended to be specific to particular conditions at geologic repositories.

TABLE OF CONTENTS

Page

Acknowledgements iii

Foreword iv

List of Tables viii

List of Figures x

B-1.0 INTRODUCTION 1

1.1 BACKGROUND AND PURPOSE OF STUDY 1

1.2 SCOPE OF TASK B EFFORT 4

B-2.0 SUMMARY 5

2.1 GENERAL 5

2.2 EVALUATION OF PROCESSING AND PACKAGING TECHNOLOGY 5

2.2.1 High-Level Liquid Wastes 6

2.2.2 Cladding Hulls and Fuel Bundle Residues 7

2.2.3 Spent Fuel Elements 8

2.3 SELECTION OF REFERENCE CASES FOR TASK D RISK EVALUATIONS 8

2.4 ANALYSIS OF ALTERNATIVE GEOLOGIC DISPOSAL TECHNIQUES 9

2.5 EFFECTIVENESS OF ENGINEERING CONTROLS 14

2.5.1 Canister Integrity 14

2.5.2 Matrix Leach Resistance 14

2.5.3 Solubility Characteristics of Leached Waste Forms 16

2.6 COST CONSIDERATIONS 16

B-3.0 ANALYSIS OF TECHNOLOGY 17

3.1 GENERAL 17

3.2 ALTERNATIVE DISPOSAL FORMS FOR HIGH-LEVEL WASTES 18

3.2.1 High-Level Liquid Wastes 18

3.2.2 Cladding Hulls and Fuel Bundle Residues 52

3.2.3 Storage Canisters 63

3.2.4 Off-Gas Treatment 66

3.2.5 Spent Fuel Assembly Disposal 70

3.3 SELECTION OF REFERENCE TECHNOLOGIES 74

TABLE OF CONTENTS (continued)

			Page
B-3.3.1		Calcination	75
	3.3.2	Glassification	75
	3.3.3	Spent Fuel Disposal	75
	3.3.4	Canisters	76
	3.3.5	Off-Gas Residue Disposal	76
	3.3.6	Cladding Hulls and Fuel Bundle Residues	77
	3.3.7	Summary of Reference Cases	78
3.4		DISPOSAL OF OTHER TRU-CONTAMINATED WASTE	79
	3.4.1	Combustibles	81
	3.4.2	Non-Combustibles	84
	3.4.3	Wet Wastes (Liquids and Sludges)	88
	3.4.4	Final Packaging	88
3.5		ALTERNATIVE GEOLOGIC DISPOSAL TECHNIQUES	88
	3.5.1	Design Characteristics for Disposal in Bedded Salt	90
	3.5.2	Disposal in Other Media	106
	3.5.3	Thermal Analysis	111
3.6		EFFECTIVENESS OF ENGINEERING CONTROLS	142
	3.6.1	Reference Technologies in Bedded Salt	142
	3.6.2	Reference Technologies in Other Media	156
B-4.0		COST CONSIDERATIONS	159
4.1		GENERAL	159
4.2		SPENT FUEL STORAGE COSTS	160
4.3		POST STORAGE/PRE-BURIAL COSTS	160
	4.3.1	Solid HLW Disposal Path	160
	4.3.2	Spent Fuel Disposal Path	164
4.4		REPOSITORY COSTS	165
	4.4.1	Costs for Geologic Disposal in Salt	165
	4.4.2	Cost Versus Depth	171
4.5		COST SUMMARY FOR HLW OR SPENT FUEL DISPOSAL IN SALT	171
4.6		COSTS FOR GEOLOGIC DISPOSAL IN HARD ROCK	171

TABLE OF CONTENTS (continued)

Page

B-4.7 NOTE ON THE DISPOSAL COSTS FOR LOW-LEVEL TRU WASTES 171

References 175

Appendices 179

Appendix B-I	Glossary and List of Abbreviations	B-I-1
Appendix B-II	Solidification Processes in Early Stages of Development	B-II-1
Appendix B-III	Off-Gas Treatment Technology	B-III-1
Appendix B-IV	Details of Reference Disposal-Site Facilities	B-IV-1
Appendix B-V	Corrosion of Metal Containers	B-V-1

LIST OF TABLES

Table No.		Page No.
B-1	Reference Cases for Various Waste Forms	10
B-2	Waste Calcine Characteristics	20
B-3	Waste Glass Characteristics	20
B-4	Principal U. S. Solidification Programs	22
B-5	Principal Foreign Solidification Programs	24
B-6	Solidification Product Characteristics	25
B-7	Characteristics of Borosilicate Glass	39
B-8	Fuel Bundle Properties	53
B-9	Fuel Assembly Disposal Summary	73
B-10	Incinerators Used in the United States for the Combustion of Radioactive Solid Wastes	85
B-11	Processes Under Development in the United States for the Combustion of Radioactive Solid Wastes	87
B-12	Volumes of Waste to be Stored in Reference Facility	93
B-13	HLW/Spent Fuel Retrievable Storage Area Data	99
B-14	Reference Facility Dimensions for TRU Waste	101
B-15	Total Facility Area Requirements	102
B-16	Conservative Estimates of Gross Vertical Expansion ("Thermal Uplift") for Different Repository Media	110
B-17	Thermal Properties of Geologic Media	115
B-18	Geologic Formations at Depths in Model	117
B-19	Parameters Affecting the Time-Dependent Heat Generation Rate	119

LIST OF TABLES (Cont'd)

Table No.		Page No.
B-20	Glass Leach Rate Constants	146
B-21	Solubility Limits of Principal Species from Actinide Dissolution	157
B-22	Approximate Costs, Reference Repository in Bedded Salt--Non-Retrievable Storage for Equivalent of 107,000 MTHM	166
B-23	Approximate Costs, Reference Repository in Bedded Salt--Fully Retrievable Storage for Equivalent of 107,000 MTHM	167
B-24	Plant Staff Estimate for Reference Repository	169
B-25	Summary of High-Level Waste Disposal Costs	170
B-26	Reference Facility Costs (Bedded Salt)--Non-Retrievable Storage of Low-Level TRU Waste	172

APPENDIX

B-III-1	Production of Carbon-14 in Light-Water Reactors	B-III-15
B-III-2	Description of Possible Flowsheet Variations	B-III-27
B-III-3	Analysis of Flowsheet Variations	B-III-28
B-V-1	Corrosion of Titanium Alloys in Sodium Chloride Solutions	B-V-3
B-V-2	Corrosion of Inconel Alloys in Sodium Chloride Solutions	B-V-6
B-V-3	Corrosion of Stainless Steels in Sodium Chloride Solutions	B-V-7
B-V-4	Corrosion of Mild Steel in Sodium Chloride Solutions	B-V-9
B-V-5	Comparison of the Corrosion Rate of Zirconium with other Alloys in Chloride Solutions	B-V-10

LIST OF FIGURES

Figure No.		Page
B-1	Fluidized-Bed Calcination	29
B-2	General Calcination Flowsheet for the NWCF	30
B-3	Continuous Inert Bed Fluidized-Bed Calciner	33
B-4	Spray Calciner for 5 MT/Day Reprocessing Facility	35
B-5	Rotary Kiln Calcination	37
B-6	In-Can Melting	40
B-7	Process Summary - Continuous Ceramic Melter	43
B-8	French Rotary Kiln - Continuous Metallic Melter	45
B-9	Original VERA Process	47
B-10	VERA Process Systems	48
B-11	Simplified Flowsheet of FINGAL Process	50
B-12	New HARVEST Pilot Plant Flow Diagram	51
B-13	Decision Areas in the Management of Fuel Bundle Residues	56
B-14	Treatment of Fuel Hulls - Process-Product Characteristics and Status	62
B-15	Typical Post-Fill Treatment of Solidifed HLLW Canisters	67
B-16	Management Methods for Transuranic-Contaminated Wastes	80
B-17	Development of Incineration Methods for Transuranic-Contaminated Wastes	82
B-18	Artist's Concept - Federal Repository	97

LIST OF FIGURES (continued)

Figure No.		Page
B-19	Geometrical Array Used for Thermal Analysis	114
B-20	Assumed Values for Thermal Conductivity vs. Temperature	116
B-21	Vertical Temperature Distribution in Typical Bedded Salt Formation	120
B-22	Vertical Temperature Distribution in Typical Granite Formation (150 kW/Acre)	121
B-23	Vertical Temperature Distribution in Typical Salt Dome	122
B-24	Maximum Mine Temperature vs. Time in Three Geologic Formations (Reprocessed Waste)	123
B-25	Maximum Mine Temperature vs. Time for 10-, 20-, and 50-Year-Old Waste in Bedded Salt	124
B-26	Maximum Mine Temperature vs. Time for 10- and 20-Year-Old Waste in Granite	125
B-27	Vertical Temperature Distrubition in Typical Granite Formation (60 kW/Acre)	126
B-28	Maximum Mine Temperature vs. Time for 10-Year-Old Waste in Bedded Salt Formation	128
B-29	Horizontal Temperature Distribution in Salt for Various Times After Burial (3-D Calculation)	129
B-30	Vertical Temperature Distribution in Salt for Various Times After Burial (3-D Calculation)	130
B-31	Isotherms in Salt Around Waste Canister Midplane (Z=0) at 2-Year Point After Burial (Q=3.95 kW/Canister)	131

LIST OF FIGURES (continued)

Figure No.		Page
B-32	Horizontal Temperature Distribution in Granite for Various Times After Burial (3-D Calculations) (150 kW/Acre)	132
B-33	Vertical Temperature Distribution in Granite for Various Times After Burial (3-D Calculation) (150 kW/Acre)	133
B-34	Isotherms in Granite Around Waste Canister Midplane (Z=0) at the 2-Year Point After Burial (Q=3.95 kW/Canister)	134
B-35	Horizontal Temperature Distribution in Granite for Various Times After Burial (3-D Calculation) (60 kW/Acre)	136
B-36	Vertical Temperature Distribution in Granite for Various Times After Burial (3-D Calculation) (60 kW/Acre)	137
B-37	Isotherms in Granite Around Waste Canister at Canister Midplane (Z=0) at 2-Year Point After Burial (Q=1.58 kW/Canister)	138
B-38	Comparison of Predicted Maximum Mine Temperature vs. Time for a One- and Two-Dimensional Analysis in Salt Dome	139
B-39	Temperature Rise vs. Radius at Canister Midplane (Z=0) for a 1100-Acre Storage Facility in a Salt Dome (Result of 2-D Analysis)	140
B-40	Maximum Vertical Uplift vs. Radius for a 1100-Acre Storage Facility in Three Geologic Formations (Results of 2-D Analysis)	141
B-41	Percent of Glass Remaining (Unleached) vs. Time, $k=10^{-6} g/cm^2$-day (Conservative Value)	151
B-42	Annual Leach Rate vs. Time, $k=10^{-6} g/cm^2$-day (Conservative Value)	152
B-43	Percent of Glass Remaining (Unleached) vs. Time, $k=10^{-8} g/cm^2$-day (Possibly Attainable Value)	153

LIST OF FIGURES (continued)

Figure No.		Page
B-44	Annual Leach Rate vs. Time, $k=10^{-8} g/cm^2$-day (Possibly Attainable Value)	154

APPENDICES

B-II-1	Supercalcine Processes	B-II-2
B-II-2	Sintering Processes	B-II-2
B-II-3	Metal Matrix Formation	B-II-4
B-II-4	Glass Ceramic Process	B-II-4
B-II-5	Ion Exchange Fixation	B-II-7
B-III-1	Schematic Diagram of the ORGDP Selective Absorption Pilot Plant	B-III-5
B-III-2	Alternative Technologies for the Management of C-14	B-III-19
B-III-3	Tritium Equilibrium Model	B-III-26
B-III-4	One Proposed Tritium Separation Scheme	B-III-30

B-1.0 INTRODUCTION

B-1.1 BACKGROUND AND PURPOSE OF STUDY

One of the major environmental concerns associated with the projected increase in nuclear power generation is the handling and disposal of radioactive waste. Highly radioactive wastes must be placed in secure repositories so that their entry into the environment can be prevented for a long period of time.

Furthermore, these wastes must be managed in a fashion that assures acceptable risk to the environment from the detrimental effects of radioactive contamination. In March 1977, the Environmental Protection Agency (EPA) contracted with Arthur D. Little, Inc., for a study to provide technical support for the development of standards for the management of high-level radioactive waste (HLW).

In the context of this study, HLW sources include (1) irradiated or "spent" nuclear fuel elements discharged from a nuclear reactor for disposal; (2) those aqueous wastes containing the bulk of the fission products, resulting from reprocessing of spent fuel for recovery and recycle of uranium and plutonium; (3) the fuel cladding and structural materials associated with the fuel. In addition, specific isotopes produced as a result of nuclear reactions associated with irradiated fuel constitute items of special interest from the viewpoint of environmental protection, e.g., carbon-14, iodine-129, krypton-85, and the longer-lived heavy elements.

The existence of HLW, mainly from defense programs, has already led to concern about temporary storage of relatively large volumes of liquid wastes, especially since there have been leaks of such wastes from underground storage tanks at the Hanford reservation, albeit with no demonstrable effects on the environment. The lack of fully-demonstrated processes and technologies for long-term disposal has also caused concern.

The problems associated with management of HLW have been recognized for some time, and considerable research and development work has been done on many aspects of these problems. Although this report will review the problems in detail, it is important to stress at the outset that considerable progress has been made toward solving them. Indeed, a preferred methodology for handling high-level wastes has evolved; this method involves isolation of solidified forms of these wastes in deep geologic repositories.

Briefly, the approach is to impose multiple barriers in series between the solidified radioactive waste material and man's environment. If the probabilities of failure for each of these barriers are small and reasonably independent, the overall probability of environmental contamination can be made quite small. The multiple barrier concept has been used successfully in other areas, e.g., control of radiation releases from nuclear power plants.

As for the barriers themselves, the principal ones are:

(1) Waste Matrix: Incorporation of the wastes into a solid matrix material resistant to attack by groundwater--the major vehicle for possible transport of wastes from the repository to the environment.

(2) Containment: Packaging of solidified wastes in a suitable container to reduce the probability of radioactive contamination during handling and during disposal operations, and to delay the onset of attack of the matrix material.

(3) Geologic Isolation: Deposit in a deep site selected for its low probability of (a) groundwater penetration and ultimate transport to the environment, (b) massive faulting or uplifts that would cause loss of isolation, and (c) accidental penetration by man.

(4) Adsorption: Reducing radioactive contamination by using an effective natural barrier. Substrate materials tend to retain many radioactive elements temporarily. Therefore, even in the event of transport by groundwater, adsorption will serve as a mechanism to decrease the transport rate of radioactive material and thereby allow time for additional radioactive decay to less-hazardous levels.

To characterize these barriers more specifically, a case being given serious consideration as a reference concept in the United States is incorporation of wastes in a glass matrix contained in a protective metal canister, buried in a deep mine located in a geologically ancient salt bed. The glass has excellent resistance to leaching by groundwater, and the ancient salt strata, by their very existence, have demonstrated long-term isolation from the surface and from contact with groundwater.

The preceding discussion has focused on geologic disposal of high-level radioactive wastes from reprocessing operations. Within the past year questions have arisen about disposal of another waste form--spent fuel elements. The question arose because of an indefinite moratorium on fuel reprocessing imposed by the present Administration, concerned over possible diversion of plutonium for weapons production. At the present time, it is not clear when reprocessing might be undertaken in the United States; there is at least a question as to whether it will ever be undertaken. If reprocessing is indeed not permitted, disposal of non-reprocessed spent fuel would be required; i.e., spent fuel would be the ultimate waste form.

In view of the very large fuel values (uranium and plutonium) remaining in spent fuel elements, many people believe it unlikely that spent fuel would actually be "thrown away" or permanently buried as waste; instead, there would be an interim storage of spent fuel elements, with ultimate reprocessing at such time as suitably stringent controls are placed on use of plutonium fuel. Nevertheless, spent fuel elements have been considered as a possible waste form to cover the possibility that the "throwaway" fuel cycle might be adopted.

Although much has already been accomplished in solving the problems of managing radioactive waste, the primary thrust to date has been in the research and development area, with only limited demonstration and prototype projects. Accordingly, the Federal Government has substantially increased the scope of its waste management programs. As part of the newly expanded program, several avenues are being intensively pursued. These include demonstration projects on engineering controls, such as fixation in glass matrices, hydrogeological and geochemical investigations to gain better understanding of potential pathways to the environment, and a detailed terminal storage design program leading to construction and operation of a disposal facility in at least two deep geologic formations.

EPA has already agreed to publish proposed generally applicable environmental standards for the ultimate disposal of radioactive waste. To help establish the technical bases for HLW standards, EPA has embarked on this technical support project.

This technical information will be used by EPA in evaluating the environmental acceptability of various options in the ERDA waste management program and of presently-operating and proposed disposal sites for high-level radioactive wastes. Furthermore, a major Environmental Impact Statement (EIS) is being prepared by ERDA to assess the total U.S. waste management program. The current schedule for issuing this EIS is during calendar year 1977. Clearly, public understanding would be enhanced if the proposed EPA environmental standards for high-level waste were available during the review phase of this major EIS.

The schedule for this study was set by the original target date of December 1977 for publication of proposed standards. According to the terms of the contract, the study is limited to reviewing and assessing published data from programs related to irretrievable deep geologic disposal that are now being conducted under ERDA and NRC sponsorship, in order to provide EPA with an independent assessment of the state of the art in management of radioactive wastes.

The effort is divided into four tasks:

Task A - Source Term Characterization/Definition
Task B - Effectiveness of Engineering Controls
Task C - Assessment of Migration Pathways
Task D - Assessment of Accidental Pathways

This report represents the results of the work done under Task B above.

B-1.2 SCOPE OF TASK B EFFORT

The scope of work for this task is divided into two major steps: (1) analysis of technology for engineering control of high-level wastes and (2) projections of costs for various alternative disposal technologies.

Most of the Task B effort has been devoted to (1), primarily because the information available for (2) is limited. The technological assessment starts with a comprehensive review of alternative disposal forms for high-level wastes, including cladding hulls, fuel bundle residues, and spent fuel elements themselves. Based on this review, reference disposal forms have been selected to span a range of isolation effectiveness. These reference cases can then be used in the Task C and D studies to test the sensitivity of environmental risk to changes in waste form and engineering controls. For example, for HLW from reprocessing plants, both calcine (high-leachability) and glass (low-leachability) matrices are considered, while for spent fuel both a high-integrity titanium canister and a low-integrity steel canister are considered.

The overall measure of effectiveness of engineering controls, as defined in Task B, is the rate at which radioactive material becomes available for further transport out of the repository. The rate of such transport depends principally upon two factors: the rate of removal from the matrix material, and the solubility of the radioactive elements in the surrounding groundwater. Data are summarized herein for each of these factors and for several alternative waste forms; these data serve as basic inputs to the Task C and Task D efforts.

Primary emphasis has been placed on high-level radioactive waste from reprocessing or present in non-reprocessed spent fuel (throwaway cycle). Brief consideration has been given to lower-level transuranic-contaminated wastes, which arise primarily from plutonium fuel fabrication and reprocessing plants. The objective of this limited review is to provide some insight into the state of the art for applying engineering controls to such wastes, including, where necessary, direct burial with high-level wastes in a geologic disposal facility.

B-2.0 SUMMARY

B-2.1 GENERAL

The effectiveness of engineering controls for isolation of high-level radioactive wastes hinges on two key factors: (1) the form of the waste material and its resistance to transport, and (2) the location and design of the geologic disposal facility to achieve maximum isolation from the environment. Item (1) includes the form of the waste product, the type of containment, the resistance of the waste matrix to leaching, and the solubility of the leached radioactive elements in groundwater since all of these affect the rate at which water might transport radioactivity from the repository. It includes certain aspects of the facility design that might conceivably influence the degree of isolation afforded by the facility itself. In particular, the engineering design of the facility has been characterized in sufficient detail to identify potential design uncertainties that might impact on the Task D risk analysis. Other aspects of facility design, such as characteristics of model sites, selection of alternative geologic media, etc., are not considered herein, but are discussed in the Task C report.

In assessing the effectiveness of engineering controls, consideration has been given in this study to a spectrum of waste forms with varying degrees of transport resistance and differing amounts of radioactive material--e.g., spent fuel with relatively higher actinide content and higher leachability versus reprocessing plant wastes incorporated in glass with lower actinide content and lower leachability. All wastes, whatever the form, are assumed to be at least ten years old when processed and placed in the repository. This assumption is consistent with the reality of today's reprocessing moratorium as well as the probability that, even if reprocessing were started, spent fuel will continue to be stored for at least a ten-year period.

The major thrust of the analysis of engineering controls has been aimed at salt deposits, because this is the only type of geologic medium for which available design data are sufficiently detailed to permit meaningful design reviews. For other media, such as shale, basalt, or granite, only limited engineering data are available, and the development of additional data is beyond the scope of this study. Detailed characterizations of repository designs have not been made for these alternative media. Instead, this review has been limited to some generalized comparisons between certain salt repository design parameters and those for other geologic media.

B-2.2 EVALUATION OF PROCESSING AND PACKAGING TECHNOLOGY

A number of possibilities have been considered for processing (solidification) and packaging (containment) of high-level wastes for permanent disposal in a Federal repository. This study has considered various HLW sources: high-level liquid wastes, cladding hulls and fuel bundle residues, and spent fuel elements. The preferred technologies for each of those waste sources will be briefly described in the following paragraphs. Possible processing and packaging approaches for low- to intermediate-level, transuranic (TRU) contaminated wastes as well as for certain other waste products from reprocessing and waste-processing plants, e.g., iodine-129, carbon-14, krypton-85, tritium, and ruthenium-106, have also been reviewed. In general, the processing problems associated with those other waste products seem solvable, although refinements may be needed in the specification of regulations for disposal of these wastes.

B-2.2.1 High-Level Liquid Wastes

High-level liquid wastes (HLLW) are generated by a spent fuel reprocessing plant. Present NRC regulations (10 CFR 50, Appendix F)require solidification of HLLW within five years after reprocessing and transfer to a Federal repository within ten years after reprocessing.

Extensive research and development effort has taken place throughout the world on possible alternatives for processing of HLLW. Some techniques have been successfully demonstrated on a near-commercial, or commercial scale; most of this work has focused on converting HLLW to solid form. In the present regulatory framework it seems unlikely that liquid waste disposal would be permitted in the near term on a large-scale commercial basis. Direct disposal of liquid wastes has therefore not been evaluated in this study.

Turning specifically to the question of solidification of HLLW, two technologies are preferred -- calcination and glassification. Calcination (roasting at high temperature) involves the production of a granular powder that is stable and less mobile than liquid wastes but still highly leachable. Extensive practical operating experience has been obtained with the fluid-bed calciner for defense wastes, but the simpler spray-calciner seems preferable for commercial wastes. (Because the spray-calciner product is a fine particulate, Battelle Northwest Laboratories (BNWL) has pointed out that a fluidized-bed calciner might actually be preferable if calcine were to be the final product.)

Glassification has a distinct advantage over calcination in that a vitrified solid with excellent leach resistance is produced. Glassification is accomplished by melting calcined waste together with glassmaking "frits" to form a relatively homogeneous solid solution of waste products in glass. The continuous ceramic melter process using resistance heating is an attractive option for glassmaking and has the advantage of having been extensively used in the glassmaking industry. An alternative process, which has been much more extensively demonstrated with simulated and actual radioactive wastes, is the in-can melter process, where the glass is formed by heating in a metal canister. A disadvantage of the in-can melter is that it subjects the canister material to higher temperatures than does the separate, pot-type, continuous melter, and therefore may require different canister materials.

For the canister itself, several materials are possible, depending upon the specific process and geologic medium. Carbon steel is suitable for the ceramic melter process, but stainless steel or Inconel is probably required for in-can melting. All of these metals have relatively high corrosion rates in a hot brine environment. For glass, this is not considered to be a serious problem because the glass itself, with its low leach rate, is really the effective containment barrier.* For calcine, however, more corrosion-resistant metals might be considered, e.g., titanium, which has excellent corrosion resistance in hot brine (see Appendix B-V).

Borosilicate glass appears to be the most suitable type of glass because of its favorable leaching characteristics and because it does not react with certain ferrous metals as do some other types of glass, e.g., phosphate glass.

Alternative solidification systems with potential for improved process or product characteristics are in various stages of development, but improvements may not be necessary or even desirable if they lead to more complex process requirements. The evidence from the technology assessment of already-developed waste solidification processes supports the conclusion that a selection could be made today from several processes that would function satisfactorily, assuming competent design, installation and operation.

B-2.2.2 Cladding Hulls and Fuel Bundle Residues

Cladding hulls and fuel bundle residues arise during mechanical chopping and eventual dissolution of fuel bundles or fuel rods at the initial stages of reprocessing. The dissolution step leaves these hulls slightly contaminated with fission products and TRU elements. From the standpoint of waste management, the preferred process is mechanical separation of relatively uncontaminated fuel-bundle hardware prior to chopping of the fuel rods and subsequent dissolution. The undissolved hulls could then be washed, compacted, and packaged in steel drums. If necessary, although it may not be cost effective, further decontamination could be accomplished or the hulls could be placed in some form of solid matrix, e.g., concrete or even glass. There is no proven technology for such packaging at present.

Selection of the best technology for disposal of cladding hulls will depend upon the evaluation of more refined guidelines for disposal of TRU-contaminated wastes. The current proposed guidelines, which have a single cut-off point of 10 nanocuries per gram (10^{-8} Ci/g), do not recognize the relative risks associated with a wide spectrum of low- to intermediate-level TRU-contaminated wastes.

---

*Recent studies(1) question the validity of this assumption at higher temperatures with water present, and may thus impose further restrictions on the allowable design temperatures within repositories.

B-2.2.3 Spent Fuel Elements

Because spent fuel elements contain approximately the same amount of fission products as HLW from reprocessed fuel, both waste forms have comparable heat generation rates and radioactivity for the first 500 years. Beyond that point, however, the higher plutonium content of spent fuel leads to higher radioactivity and greater heat generation.

The simplest disposal method for spent fuel would be containment in an outer steel canister purged and backfilled with dry nitrogen and then seal-welded. Depending upon the leach rate for spent fuel elements, a quantity that cannot be estimated now with any accuracy, it may be worthwhile to consider a high-integrity containment, such as titanium, for spent fuel elements. Corrosion rate data for titanium in hot brine indicate that containment lifetimes of the order of 500-1000 years may be obtainable.

Some further elaboration on spent fuel leach rates is appropriate. The principal uncertainty concerns the chemical environment after the containment and Zircaloy cladding have corroded away. On the one hand, in an oxidizing environment at 200°C, UO_2 rapidly oxidizes to U_3O_8; corresponding physical changes cause the dense ceramic material to disintegrate, leading to extremely high surface/mass ratios and, hence, high leach rates. On the other hand in a reducing environment, it is possible that the UO_2 could retain its ceramic form; some laboratory tests show very low leach rates for UO_2 under these conditions. Much more data and evaluation are needed to characterize spent fuel leach rates properly.

B-2.3 SELECTION OF REFERENCE CASES FOR TASK D RISK EVALUATIONS

Given the wide range of possibilities, the varied state of technological development, and the differing degrees of effectiveness for the alternatives, some weeding out of less suitable alternatives is essential, in order to focus the risk analysis effort (Task D) on the most suitable techniques.

The approach used for this selection process has been to choose a set of reference cases that span a reasonable range of possibilities in terms of (a) waste form, (b) processing (solidification) approach, (c) packaging (containment) method, and (d) isolation effectiveness. In selecting reference cases, an attempt has been made to span a wide enough range of possibilities to permit interpolation and/or extrapolation to other combinations of variables not specifically included in the reference cases.

Additional criteria used in selection of reference technologies for treatment and disposal of HLLW are as follows:

1. The reference case waste characteristics should span the reasonable range of possiblities available in the near term (within ten years).

2. The reference case technologies should be available for full-scale deployment for treatment of reprocessing wastes.

3. Reference cases are based on U.S. technology, since there are several processes for solidification available in the United States.

Choice of the following three reference waste forms for HLW seems appropriate: calcine, borosilicate glass, and spent fuel. (These span a reasonable range of possibilities in terms of likely waste forms and leach resistance.) For containment, calcine wastes are assumed to be placed in carbon steel or titanium canisters, to bracket the range of containment effectiveness. For glass, a stainless steel canister is assumed (to accommodate in-can melting process temperatures). Higher-integrity containment (e.g., titanium) was not considered because of the inherently high leach resistance of glass. Spent fuel is assumed to be placed in either carbon steel or titanium canisters with dry nitrogen gas backfill. Because spent fuel could be as leachable as calcine, these two canister types provide a further test of the effect of canister corrosion rates on transport of radioactive material out of the repository.

For cladding hulls and fuel bundle residues, the reference case is based on available technology: mechanical removal of fuel bundle hardware from clad fuel rods, compaction of chopped hulls, and packaging in carbon steel canisters.

Reference cases have also been selected for other waste forms; these will be described in detail in the body of the report. Table B-1 summarizes all the reference cases for various waste forms.

B-2.4 ANALYSIS OF ALTERNATIVE GEOLOGIC DISPOSAL TECHNIQUES

Disposal of radioactive wastes in deep, stable geologic formations has long been considered the preferred method for isolation of these wastes from contact with man's environment. A number of possible geologic media have been considered for such disposal, including salt beds, salt domes, crystalline rock forms such as granite or basalt, shales, limestones, certain types of clay beds, and others. To date, salt deposits have received the most attention, especially in the United States, because of their demonstrated natural stability over extremely long time periods.

TABLE B-1

REFERENCE CASES FOR VARIOUS WASTE FORMS

Source of Waste	Converted Waste Form	Assumed Mode of Disposal	Canister Material	Notes
HLLW	calcine	deep geologic burial	(a) carbon steel	spray calcination process
			(b) titanium	spray calcination process
	borosilicate glass	deep geologic burial	stainless steel	in-can melt process
Spent Fuel	none	deep geologic burial	(a) carbon steel	dry nitrogen backfill
			(b) titanium	dry nitrogen backfill
Cladding Hulls*	none	deep geologic burial	carbon steel	compacted to 1/3 original volume
Krypton-85	none	storage at reprocessing plant	steel (pressure cylinders)	stored for ∿100 years for decay
Iodine-129	none	deep geologic burial	carbon steel	as silver zeolite and mercuric iodate
Carbon-14	none	deep geologic burial	carbon steel	as calcium carbonate
Tritium		release		

---

*For this study fuel bundle hardware is assumed to be separated from cladding hulls and treated as low-level waste.

Moreover, salt deposits are homogeneous and are capable of plastic flow. The self-healing characteristics of salt effectively eliminate the possibility of extensive cracking, thereby preventing the opening up of pathways to the environment. For salt to remain intact, it must continue to remain dry, and thus must be protected from uncontrolled drilling activities.

An alternative to salt is a stable crystalline rock such as basalt or granite. There are abundant examples of such rock of suitable depth and age with demonstrated seismic stability. Crystalline rock does not have the self-healing characteristics of salt, but possesses other advantages, e.g., resistance to water intrusion, that make it a desirable medium for geologic disposal of radioactive waste.

In addition to salt and crystalline rock, other geologic media have been considered, e.g., shales and clay deposits. In general, these have both desirable and undesirable characteristics, depending upon the specific medium and type of waste. For example, the laminar structure of horizontally-bedded shale reduces water permeability in the normal (vertical) direction but the presence of water in shale could lead to high stresses and possible disintegration if the shale is subjected to high temperatures. In a similar vein, certain types of clay-till deposits have the advantage of low water permeability, but the disadvantage of indeterminate long-term stability characteristics.

In light of the foregoing considerations, the following cases have been selected as reference cases for the purposes of this study:

Case 1. Disposal in bedded salt.
Case 2. Disposal in granite.
Case 3. Disposal in salt domes.

A uniform depth of 460 meters (1500 ft) has been assumed for all three cases.

Until recently, nearly all of the work performed on engineering design of disposal facilities has been for salt deposits; only limited information is available for media other than salt. Therefore, the major focus of this report is on salt deposits.

The design of waste disposal facilities generally follows conventional mining practices, with two important exceptions: (1) special provisions are required for safe transport and cooling of radioactive material; and (2) waste canister spacing and arrangement must be in accordance with thermal design criteria for the specific medium. The thermal criteria are basic to the repository design.

The radioactive wastes release heat to the repository at a rate that decreases with time. In the earlier years, heat is not transferred out of the repository as fast as it is generated and the stored thermal energy causes repository temperatures to rise. As temperatures rise, heat begins to flow to the surface, but it is a few hundred years before the heat flow from the surface to the atmosphere significantly influences the temperatures in a deep repository. Ultimately, as heat generation rates decline, heat losses to the surface exceed heat generation rates and repository temperatures go down. Elevated temperatures are present for about 1000-10,000 years, depending upon the medium, the repository depth and the overall heat generation rates; peak temperatures are reached in 100-500 years for a 460-m (1500-ft) deep repository. Basically, the problem can be simply summarized as a heat storage phenomenon concurrent with slow transfer of heat to the surface, and the temperature and temperature gradients must be limited to values that do not have an adverse impact on the integrity of the repository. The actual limits depend upon the type of geologic medium, e.g., salt versus granite.

The only way to limit aggregate (as opposed to localized) repository temperatures is to control the amount of heat produced per unit of horizontal area over an extended time period, since most of the heat transfer must occur in the vertical direction through a thickness that is large compared with that of the repository itself. The waste heat output is described per unit time and per unit horizontal area at some fixed point in time; it is called "planar heat density" in this report. The usual units are in kilowatts per acre and, unless otherwise noted, all values in this report are for ten-year-old wastes. (Historically, values of about 150 kW/acre have been used in the engineering designs considered for salt repositories at 305-m (1000-ft) depth.) Clearly, the planar heat density is significant, since the size of the mined area for HLW disposal is inversely proportional to planar heat density.

The allowable planar heat density may depend upon such considerations as allowable temperatures within the waste canisters, in the salt or rock adjacent to the canisters, throughout the mined area, in neighboring aquifers, on the surface, and in the ground outside the repository boundaries. These factors may also limit the allowable vertical uplift arising from thermal expansion of the entire repository. These thermal considerations are discussed in detail within this report; in conjunction with other engineering data on waste canister dimensions, shielding requirements, etc., they are sufficient to permit preliminary design of repositories in salt deposits. Considerable conceptual design work has already been performed under the direction of ERDA's Office of Waste Isolation for disposal of HLW in bedded salt. The reference facility design, reviewed in detail herein, is assumed to be located in a salt bed at 460-m depth and with a thickness of 50-100 m. The mined area is roughly a square 3000 m on a side, covering an area of about 2000 acres. It is large enough to store all wastes produced over the lifetime of 140 GW of nuclear plant capacity. For an assumed addition of 700 GW to total Light Water Reactor (LWR) capacity (see Task A report), five such facilities would eventually be required, but these need not necessarily be on separate sites.

A detailed description of the facility design will not be given here, but some of its important physical characteristics will be noted. About 50% of the area is for low- and intermediate-level TRU wastes and the remainder is for HLW canisters. A system of tunnels is used to ensure adequate support for overburden and to maintain the proper spacing of individual cylindrical HLW canisters, each 30.5 cm in diameter by about 3 m high. Canisters are transported in shielded casks and/or shielded transport vehicles during handling. Holes are drilled in the tunnel floors, waste canisters are inserted, and the holes are backfilled with crushed salt. The assumed reference planar heat density (over the HLW field) is 126 kW/acre for ten-year-old waste. Additional details on the preliminary mine design and proposed operating procedures for HLW disposal may be found in the body of this report.

No detailed engineering work has been reported on facilities for spent fuel disposal. Because of higher long-term heat generation rates, disposal of spent fuel may require lower planar heat densities, roughly 40-50% of the value for HLW. The existing conceptual design for an HLW facility has been modified to accommodate spent fuel. Since the energy output of a spent fuel bundle in an individual canister is about 1/9th that of an HLW canister, if the planar heat density must be reduced by a factor of 2-2.5, then the number of spent fuel canisters per unit area should be roughly four times that for HLW canisters. This study has assumed that four spent fuel canisters would be buried in one hole, with the holes on approximately the same grid spacing as for HLW canisters. This would result in a 150% increase in the size of the mined area, but this increase is partially offset by a substantial reduction in the area required for TRU wastes. The net effect is an increase of about 75% (from 2000 to 3500 acres) for the mined area.

The preceding discussion has focused on disposal in salt deposits. Because of the lack of detailed engineering data, it has not been possible to make a thorough assessment of disposal technology in granite or shale. A thermal analysis of granite repositories (presented herein) coupled with limited engineering studies supported by Atomic Energy of Canada Limited (AECL), suggest that granite repositories are feasible, with lower planar heat densities than in salt. Changes in canister size and/or heat loading could also be required for granite repositories. The need for these changes depends on the limits for cracking and/or spalling (from thermal stresses) and for vertical uplift (from thermal expansion), neither of which have yet been developed. Based on the AECL work, it has been assumed that a granite repository would be roughly twice the size of a comparable repository in bedded salt.

There appear to be insufficient data to predict the feasibility of repositories in rock other than granite.

B-2.5 EFFECTIVENESS OF ENGINEERING CONTROLS

The technology for design of geologic disposal facilities has been summarized in the preceding section. This section presents a review of available data on effectiveness of the three major barriers to transport radioactive waste from the repository: canister integrity, matrix leach resistance, and solubility characteristics of leached waste forms.

B-2.5.1 Canister Integrity

We assume corrosion to be much more of a threat to canister integrity than stress caused by rock or salt movement in the short term. Canister lifetimes can range from as little as several years to more than 1000 years, depending upon canister material and repository characteristics. Long-term canister integrity may be important only for waste forms with relatively high leach rates, i.e., calcine and, possibly, spent fuel elements. The use of high-integrity canisters with expected lifetimes up to 1000 years appears to be feasible through use of thicker sections of conventional materials (carbon/stainless steels) or, in salt, through use of a material, perhaps titanium, with a low corrosion rate. The technology is available for fabrication of the required canister types, but use of titanium would substantially increase canister costs.

B-2.5.2 Matrix Leach Resistance

Three types of matrices were considered--glass, calcine, and spent fuel material.

(a) Glass -- Although many years of laboratory work on glass leaching show it to be a very slow phenomenon, the theoretical models and physical data are as yet conflicting, imprecise, and scanty. The effects of temperature, flow, pH, and salinity have not even begun to be studied in an organized way for the spectrum of suitable glasses. Moreover, the extrapolation of short-term laboratory tests to very long time periods is made difficult by the limited amount of long-term leaching data. It is possible that short-term leach rate data substantially overstate actual long-term leach rates.

On the other hand, there are also concerns about the stability of a glass matrix over the very long term. There is considerable evidence that certain types of man-made glass last for several thousand years and some natural glasses are much older. One obvious difference between these glasses and HLW glass is the presence of radiation effects in the latter. Although accelerated life testing has indicated no significant devitrification, concern about radiation damage persists, since glass lifetimes beyond 10,000 years probably cannot ever be demonstrated conclusively, even with accelerated life testing. Recent work at Pennsylvania State University(1) has indicated rapid rates of glass alteration at hydrothermal conditions (300°C, 300 atmospheres) that appear to be more severe than would be likely in a properly designed repository. These findings have not yet been applied in the design of an entire repository system.

Given the uncertainties in the physical data, along with the continuing and essentially unanswerable concerns over long-term deterioration, it appears unreasonable to do more than assume a conservatively high value for glass leach rates--expressed in terms of the bulk glass corrosion rate (g of glass per cm^2-day). A value of 10^{-6} g/cm^2-day has been chosen for this study because it is at least a factor of 7 and as much as a factor of 100 larger than any reported data at the one-year point. By assuming no decrease with time, further conservatism has been introduced, which should probably more than compensate for any uncertainties concerning long-term deterioration, e.g., devitrification. At the same time, some experimental data suggest that much smaller leach rates may occur in practice. These might be in the range of 10^{-8} g/cm^2-day or even less. Accordingly, a leach rate of 10^{-6} g/cm^2-day has been adopted as a reasonably conservative long-term average value, and 10^{-8} g/cm^2-day as a possibly achievable long-term average value, assuming that the severe conditions of the Pennsylvania State University work are not approached.

To determine the total release rate of radioactive material, glass leach rates (g/cm^2-day) must be combined with surface-to-mass ratios (cm^2/g) to give fractional release per day. An initial surface-to-mass ratio of about 0.4 cm^2/g has been assumed, consistent with test data on actual canisters after normal cooling transients and additional breakage from normal handling.

The resulting initial release rates (% of initial mass) range from roughly 0.01%/year down to 0.0001%/year, depending upon the assumed value of the bulk glass leach rate constant. For the high leach rate, one-half the glass is dissolved after about 4000 years (with total dissolution after 20,000 years), while the corresponding figures for the low leach rate are 400,000 years and 2 million years.

(b) Calcine--Calcines are relatively soluble and have high surface-to-mass ratios; hence, full dissolution is expected to occur in less than one year. Such high release rates are of special concern during the first few hundred years because of undecayed fission product radioactivity. In this time period, the canister, not the matrix, would be the primary barrier.

(c) Spent Fuel--As already noted in Section B-2.2.3, leach rates for spent fuel elements are highly speculative. In particular, the leach rate for the UO_2 fuel material can vary over a wide range, depending upon temperature, groundwater chemistry, and oxygen content. The range of possible release rates spans several orders of magnitude, from a value as high as 10^{-1} g/cm^2-day to a value of about 10^{-6} g/cm^2-day. Considerably more effort is required to reduce the uncertainty band in spent-fuel leach rates, especially with respect to the probability of the presence of oxygen-rich groundwater which would cause rapid deterioration of the UO_2 ceramic material.

Given the large uncertainty in spent-fuel leach rates, the simplifying assumption has been made that spent fuel could have one of two possible total release rates: (1) a value equal to that of calcine, and (2) a value equal to that of high-leach-rate glass.

B-2.5.3 Solubility Characteristics of Leached Waste Forms

In cases where water flow through the repository is low, relatively high concentrations of certain isotopes in the leachate may be calculated from glass dissolution rates. A correction has been supplied to allow for the limited solubility of the actinides in the leachate. In low-flow situations this solubility may limit the rate at which actinides leave the repository.

B-2.6 COST CONSIDERATIONS

Data have been developed to indicate comparative costs for the various reference cases chosen; these data will be needed in order to evaluate subsequently the cost effectiveness of alternative disposal techniques.

Since only limited amounts of cost data have been published, and because the scope of this study did not call for making independent cost estimates, the resulting cost data should be considered only as rough estimates. Nevertheless, they provide a useful perspective on the relative costs of various disposal techniques.

By far the largest single contribution to waste-handling costs is that of interim spent-fuel storage, which, at \$80-\$150/kg HM*, can amount to roughly 8-15% of total fuel cycle costs. Other disposal costs for solidified HLW are estimated to range from \$30-\$70/kg HM, and for spent fuel are estimated to range from \$40-\$75/kg HM, because of higher shipping, canister, and burial costs.

Differences between calcine and glass solidification costs appear small in comparison with the other uncertainties in waste disposal costs.

---

*HM = "heavy metal" (U and Pu) fed to reactors.

B-3.0 ANALYSIS OF TECHNOLOGY

B-3.1 GENERAL

This section of the report deals with the various technological aspects of engineering controls to reduce the probability of release of radioactivity to the environment. These aspects include (1) the form of waste material, and (2) the design of the geologic disposal facility. Various technologies are evaluated for encapsulating and containing different types of high-level radioactive wastes. For geologic disposal, primary emphasis is on bedded salt, since this medium is the only one for which extensive information is available; a review of available data on other geologic media is also presented, in order to indicate some of the difficulties arising from insufficient data. In general, the available data do not permit detailed evaluation of the effectiveness of engineering controls for media other than salt.

Before proceeding to the detailed technology assessment, another general point should be noted concerning the assumed "age" of waste material at the time of geologic disposal. In the past, it has generally been assumed that fuel would be reprocessed relatively quickly (within 6-12 months of discharge) and that wastes would be solidified 1-5 years after reprocessing. In light of today's indefinite moratorium on reprocessing, however, it now appears that wastes will "age" as spent fuel for ten years or more until reprocessing is initiated or a decision is made to "throw away" the spent fuel by burial in a geologic repository. In either case, a substantial amount of storage capacity for spent fuel is required.

Having made such an investment in storage capacity, the industry will probably use the storage facilities for a long time. Moreover, substantial economies may be effected in the design of reprocessing plants and/or waste processing and disposal facilities with such extended storage. These economies result from the reduced heat generation and radioactivity of the aged material after radioactive decay. As already shown in the Task A report, heat generation and radioactivity decrease by roughly a factor of 3.4 between 2-year and 10-year decay times and by an additional factor of 1.6 between 10-year and 30-year decay times. Krypton-85 and tritium levels fall by factors of two or more during such storage, as well.

Perhaps even more important than the potential cost savings are the simplifications that can be made in waste processing if only aged wastes are processed (those at least ten years old). The problem of heat transfer out of vitrified matrices is greatly reduced, eliminating the need for elaborate provisions for heat transfer, such as the composite-metal matrix. Concerns over glass devitrification are also reduced because of low glass temperatures. Finally, the entire handling sequence can be made simpler, and therefore more reliable, because of reduced radioactivity and lower heat-generation rates.

In view of these advantages, the reference cases are based on the assumption that only aged wastes would be processed and buried in a geologic disposal facility.

B-3.2 ALTERNATIVE DISPOSAL FORMS FOR HIGH-LEVEL WASTES

This section of the report describes the programs and processes used to prepare (solidify) and package high-level wastes for permanent disposal in a Federal repository. The following sections cover high-level liquid wastes (HLLW), cladding hulls, fuel bundle residues, and spent fuel elements. In this section, repeated reference is made to ERDA 76-43(2), currently the most comprehensive document on the subject. Other sources are also used, but the exhaustive, thorough nature of ERDA 76-43 makes it an indispensable component of the discussion that follows.

B-3.2.1 High-Level Liquid Wastes

High-level liquid wastes (HLLW) are generated by a spent fuel reprocessing plant. These wastes are the highly radioactive solutions that remain after recovery of uranium and plutonium for reuse in the nuclear fuel cycle. Reprocessing of spent fuels from the commercial nuclear power industry is not being done in the United States at present, but a substantial amount of the HLLW from past reprocessing of defense materials is on hand. The Task A report discusses the present and probable future amounts and character of HLLW. Present regulations require solidification of these wastes within five years after reprocessing, and transfer of the solids to a Federal repository within ten years after reprocessing. As discussed elsewhere in this report, there may be economic and other advantages to delaying the reprocessing of spent fuel until at least ten years have elapsed since the discharge of the fuel from the reactor. This would substantially reduce the radioactivity (and therefore the heat removal problem) of the wastes.

Before solidification, the high-level liquid waste from commercial sources, generated at a rate of about 5000 liters/metric ton of uranium (MTU) reprocessed, will be concentrated to a liquid volume of about 380 liters per MTU.(2) This concentration would be achieved by evaporation, the purpose of which is to concentrate the HLLW and recycle water and nitric acid. The heat generated by the HLLW is a function of reactor specific power level, burnup, and time after discharge. Fuel irradiated at a power level of 38.4 MW_t/MTU to a burnup of 33,000 MW_t-days/MTU is considered typical of the LWR fuel that will be reprocessed in the United States in the next two or three decades. The rate of decay heat generated in such a fuel or in the HLLW made therefrom would be as follows:(2)

Time out of reactor	Approximate decay heat rate kW/MTU charged to reactor
160 days	24
1 year	12
10 years	1.1

Extensive research and development has taken place in the United States and throughout the world on possible alternatives for treatment of nuclear high-level wastes. Practical application of some of these techniques on a commercial or near-commercial scale has been successfully demonstrated. The results and status of these projects will be summarized in this section. It should be borne in mind that, since the wastes arising from commercial reprocessing will be "younger" and more concentrated than those from the nation's defense waste programs, the measures discussed in this report should be more than adequate for disposal of defense wastes from which the sodium salts have been removed.

The possibilities for some form of direct disposal of liquid wastes have been studied extensively. In the present regulatory framework, however, it appears unlikely that such techniques would be seriously considered for commercial application. Handling of liquid wastes presents safety problems for transportation if the waste disposal area is off the reprocessing site. Moreover, it leaves the waste in a more readily dispersible form in the near term, when the hazard is greater. Although direct disposal of liquids may eventually prove to be acceptable under some circumstances, especially if transportation is not involved, it has not been considered in this study.

Although waste solidification appears desirable, this solid waste may take a number of forms. In early practical demonstrations liquid waste was converted to a calcine by roasting at high temperatures in an oxidizing environment. Lately, work has centered on incorporation of the waste into a glass, which has substantially improved characteristics of leach rate, thermal conductivity, resistance to dispersal during transportation or handling accidents, and fire resistance. Most major programs in other countries (France, England, USSR, Germany, India, and Italy) favor glass for the final form prior to disposal.

Several research projects are studying waste forms even more durable than glass, e.g., glass-ceramics. Better-than-glass forms may be beyond the point of diminishing returns on improved product form, however.(3) Presently both calcine and glass are considered to be acceptable forms of solidification of high-level liquid wastes.(2) Typical waste calcine characteristics from acid HLLW, such as from commercial reprocessing, are shown in Table B-2.(4) Typical waste-glass properties from similar HLLW are shown in Table B-3.(4)

TABLE B-2

TYPICAL WASTE CALCINE CHARACTERISTICS

Appearance	Friable granules, scale or powder
Bulk density g/cm^3	0.7 to 2.0
Leach rate at 25°C distilled water, g/cm^2-day	10^{-1}
Thermal conductivity, W/m°C	0.2 to 0.4
Nitrate content, % by wt	0.05 to 10
Water content, % by wt	0.01 to 0.2
Thermal and radiation stability	Requires post-calcination treatment to remove nitrate and water (at 900°C)

TABLE B-3

TYPICAL WASTE GLASS CHARACTERISTICS

Appearance	Black, shiny with conchoidal fractures
Bulk density, g/cm^3	3.2 to 3.4
Leach rate at 25°C, distilled water, g/cm^2-day	10^{-8} to 10^{-6}
Thermal conductivity, W/m°C	0.8 to 1.4
Formation temperature, °C	1050 to 1150
Maximum storage temperature, °C	< 300 (adjusted for 10-year-old wastes)

The specific values of the calcine and glass characteristics are dependent on the process, the operating parameters employed, and the waste composition.(4) Therefore, the characteristics given tend to vary somewhat from reference to reference, but the above values appear to be typical. It should be noted that the calcines have relatively low thermal conductivity and high leachability; they contain residual water and nitrate, but in commercial calcining processes, these are typically baked out at about 900°C.

In the conversion of calcine to glass, the addition of glass-forming additives increases the weight of the resultant product by a factor of 2.5-3.5. The density also increases, resulting in a net product volume increase of only about 50%. As Tables B-2 and B-3 show, the leach rate for glass is much lower than for calcine. The thermal conductivity is increased by a factor of 3-5 (allowing the use of larger-diameter waste canisters, assuming similar centerline temperature limits for glass and calcine products). With its lower leach rate and higher thermal conductivity, glass is therefore generally considered to be a better waste form.

The solidified waste, either a calcine or a glass, will probably be contained in a metal canister; metals considered for this application are: carbon steel, stainless steel, Inconel, and titanium. Choice of canister type will be discussed in more detail in Section B-3.2.3.

The remainder of this section, B-3.2.1, covers the solidification of HLLW to calcines and to glass. The calcination and glassification programs that are in the most advanced stage of development, i.e., most nearly ready for commercial application, will be described first. Discussion of several processes that are in earlier stages of development will follow.

The emphasis is on the conversion of HLLW to solid forms suitable for final disposal. Other associated operations, such as off-gas treatment and inspection and handling of waste canisters, are covered in separate sections.

B-3.2.1.1 Solidification Technology Under Development

For the reader not interested in process detail, Tables B-4 and B-5 summarize the status of the principal U.S. and foreign solidification programs in an advanced stage of development. Each of the programs is discussed at more length in the subsequent sections. The characteristics of the products of all these solidification processes are shown in Table B-6.

Processes that are still in an early stage of development are discussed in Appendix B-II.

TABLE B-4

PROCESSING/TECHNOLOGY SUMMARY, DEVELOPED U.S. SOLIDIFICATION PROCESSES

	OPERATING TEMPERATURE (°C)	HEAT CONTROL	LEVEL OF DEMONSTRATION	COMMENTS AND POTENTIAL OR UNRESOLVED PROBLEMS
Fluidized Inert Bed	500-800*	In-Bed Combustion (kerosene, O_2)	INEL application at 400-500 liters/hr (military wastes). BNWL inert bed tests (non-radioactive): 38 runs 350 hr, up to 40 liters/hr (25 liters/hr nominal).	INEL process difficult to adapt to commercial wastes. Tests show inert material precludes bed melt in event of bed collapse. Sodium limit of 1.2 M without additives. No data on ruthenium. No nozzles. Conceptually more complex than spray calciner.
Spray Calcination	700-800* (wall)	Electric Resistance Furnace	Development since 1959. 13 fully-radioactive engineering scale runs (WSEP), solidifying 8700 liters in 600 hrs. Current WFP: development unit 44 runs 875 hrs (non-radioactive); full scale unit 3 runs at 210 liters/hr (non-radioactive).	1-2% of ruthenium offgassed, sodium retention up to 2 M. Requires vibrator to reduce wall scale. Early difficulties with spray nozzles appear to be corrected. No on-line commercial experience.
In-Can Melting	1000-1100	Zone Furnace	7.5 kg/hr radioactive; 50 kg/hr non-radioactive; 22 runs 2500 kg	Canisters subjected to high temperatures; possible materials limits. Possible difficulties measuring temperatures/levels in canisters (new canister used for each melt.) Temperature restraints may result in crystalline phases in glass.

---

*Followed by 900°C bakeout of water and nitrates.

TABLE B-4
(continued)

PROCESSING/TECHNOLOGY SUMMARY, DEVELOPED U.S. SOLIDIFICATION PROCESSES

	OPERATING TEMPERATURE (°C)	HEAT CONTROL	LEVEL OF DEMONSTRATION	COMMENTS AND POTENTIAL OR UNRESOLVED PROBLEMS
Continuous Ceramic Melter	1100-1200	Electrodes; Startup Using NaOH	Non-radioactive: 23 months, 4000 kg, 80 liters/hr. Tilt-pour demonstrated.	Direct feed of liquid waste may be possible; transfer of molten glass requires more equipment and higher risk of maintenance. Not tested radioactive.

TABLE B-5

PROCESSING/TECHNOLOGY SUMMARY, DEVELOPED FOREIGN SOLIDIFICATION PROCESSES

	Operating Temperature (°C)	Heat Control	Level of Demonstration	Comments & Potential or Unresolved Problems
Foreign				
French Rotary Kiln Calciner	500	Zoned external resistance furnace	5000 hrs pilot and engineering scale (non-radioactive). No radioactive experience; startup of commercial unit scheduled this year.	Potential maintenance problems due to rotating machinery (bearings and seals). No radioactive tests.
French Rotary Kiln with Continuous Melter	1150 (melter)	Induction heater (melter)	(see item above for calciner). Tests of PIVER process (similar) for 4 yrs (radioactive), 164 runs non-radioactive pilot tests of new process.	(see also item above). Freeze valve for liquid transfer may create reliability problem (plugging or unexpected valve opening); PIVER had frequent vessel failures.
German Spray Calciner with Continuous Melter	600 (calciner) 1150-1200 (melter)	Superheated steam or wall heater for calciner, induction or joule heater for melter.	Pilot scale (non-radioactive) 1000 hrs, 1500 kg.	Front-end denitration minimizes production of non-condensable gases. Use of superheated steam complicated. Freeze valve potential maintenance problem; spray nozzle used (potential plugging).
British Rising Level Glass (HARVEST)	1000-1050	Multi-zone furnace	Similar FINGAL process tested in 8 radioactive production runs, 64 non-radioactive runs. 13.8 liters/hr non-radioactive HARVEST test.	An in-can process, and discussion of in-can melting in Table B-4 applies. Eliminates calciner.

TABLE B-6

SOLIDIFICATION PRODUCT CHARACTERISTICS

PROCESS	PRODUCT	PARTICLE DIAMETER µm	BULK DENSITY g/cm^3	SURFACE TO MASS RATIO cm^2/g	LEACHABILITY g/cm^2-day	THERMAL CONDUCTIVITY W/m°C
A. DEVELOPED SOLIDIFICATION PROGRAMS IN U.S.						
Fluidized Bed	Calcine	100-300	2-2.4	10^3-10^4	HIGH	0.18-0.33
Spray Calcination	Calcine	2-5	0.5-1.3	$\sim 10^5$	HIGH	0.18-0.33
In-Can Melting	Borosilicate Glass		3.0-3.6	0.05-0.5	LOW	0.9-1.3
Continuous Ceramic Melter	Borosilicate Glass		3.0-3.6	0.05-0.5	LOW	0.9-1.3
B. DEVELOPED SOLIDIFICATION PROCESSES IN FOREIGN COUNTRIES						
French Rotary Kiln Calciner	Calcine	2-5	1-1.3	$\sim 10^5$	HIGH	0.18-0.33
French Rotary Kiln Continuous Metallic or Ceramic Melter	Borosilicate Glass		3.0-3.6	0.05-0.5	10^{-7}	0.9-1.3
German Spray Calciner Continuous Melter	Borosilicate Glass		2.9-3.1	0.05-0.5	10^{-6}	1.2-1.3
British Rising Level Glass	Borosilicate Glass		3.0-3.6	0.05-0.05	LOW	1.2-1.3
C. U.S. PROCESSES IN EARLY STAGES OF DEVELOPMENT						
Supercalcine	Crystalline Ceramic				LOW	

TABLE B-6

(continued)

SOLIDIFICATION PRODUCT CHARACTERISTICS

PROCESS	PRODUCT	PARTICLE DIAMETER μm	BULK DENSITY g/cm^3	SURFACE TO MASS RATIO cm^2/g	LEACHABILITY g/cm^2-day	THERMAL CONDUCTIVITY W/m°C
C. U.S. PROCESSES IN EARLY STAGES OF DEVELOPMENT (continued)						
Sintering	Glass-Crystalline Phase		2.4-3.3		about 10^{-8}	about 0.7
Metal Matrices	Waste Particles in Metal				Depends on waste form in metal	5-35
Glass-Ceramic	Crystal Plus Residual Glass Phase		2.85		LOW	2.2
Coated-Pellet	Coated Oxides in Metal Matrix				VERY LOW	Depends on Metal Selected
Ion Exchange	Ceramic		more than 4.5		LOW	

TABLE B-6
(continued)
SOLIDIFICATION PRODUCT CHARACTERISTICS

PROCESS	PRODUCT	PARTICLE DIAMETER μm	BULK DENSITY g/cm^3	SURFACE TO MASS RATIO cm^2/g	LEACHABILITY g/cm^2-day	THERMAL CONDUCTIVITY W/m°C
D. FOREIGN PROCESSES IN EARLY STAGES OF DEVELOPMENT (continued)						
Julich Borosilicate	Borosilicate Glass		2.9-3.1		3.6×10^{-5}	0.9-1.3
Pamela	Phosphate Glass Granules in Lead Alloy				5×10^{-7}	24-35
Thermite	Ceramic-Metal					

When no number appears, data are either not available or not applicable. In general, a "high" leachability means $>10^{-4}$ g/cm^2-day and a "low" leachability means $<10^{-7}$ g/cm^2-day.

B-3.2.1.1.1 Calcination

B-3.2.1.1.1.1 Fluidized-Bed Calcination

Fluidized-bed calcination has the longest history of operation of any technology for radioactive waste solidification in the United States. It was developed at the Idaho National Engineering Laboratory (INEL). The first engineering-scale facility for the solidification of liquid radioactive wastes was the Waste Calcining Facility (WCF), which began calcinating wastes from the Idaho Chemical Processing Plant (ICPP) in 1963. The fluidized-bed calcination process was originally developed for the acidic aluminum nitrate solutions arising from the processing of uranium-aluminum alloy fuels and was intended to be only a demonstration project. The WCF plant, however, became an integral part of the waste-management program at ICPP and it has been used to solidify aluminum nitrate, zirconium fluoride, and stainless steel sulfate waste solutions.(5) Fluidized-bed calcination has operated successfully at ICPP for about 14 years, resulting in calcination of 10.2 million liters (2.7 million gallons) of waste at production rates up to 500 liters/hr.

Calcining by the fluidized-bed method is accomplished by pneumatically atomizing the waste solution into a bed of fluidized solid granules at a temperature of 500°C. Heat is supplied by in-bed combustion of kerosene with oxygen. See Figure B-1 for a schematic representation of such a unit.

The original facility used a liquid-metal (NaK) heat-transfer system that worked adequately for the first three processing campaigns during which the system operated at temperatures greater than 600°C for 35,000 hours.(5) The plant capacity was then increased by providing a more thermally-efficient in-bed combustion system. The in-bed combustion system also has the apparent advantage of reducing the amount of ruthenium that leaves the calciner when processing zirconium wastes containing fluorides. Possibly unburned hydrocarbons or carbon monoxide reduce the volatilized ruthenium to a non-volatile lower valence state.(6) See Figure B-2 for a schematic diagram of the off-gas handling system.

The fluidized-bed calciner converts liquid waste to a mixture of powdery solids and granules ranging from 0.1-0.3 mm in size. Density varies from 2.0-2.4 g/cm^3 and the specific volume is about 40 liters/MTU for commercial reprocessing such as that originally planned for the Allied-Gulf Nuclear Services Plant at Barnwell, SC (AGNS). The in-bed combustion temperature is 500-800°C, and the calcine product can be stabilized (denitrated and dehydrated) at about 900°C for storage, or the calcine can be fed directly from the calciner to a glass melter. Thermal conductivity of the product varies from 0.18 W/m°C at 40°C to 0.33 W/m°C at 600°C.

The capacity of a fluidized-bed waste calciner is dependent on the process conditions chosen and on the particular waste being solidified. Data obtained at the WCF indicate that a 75-cm diameter calciner could accommodate the feed from a 5 MTU/day plant. The ICPP fluidized-

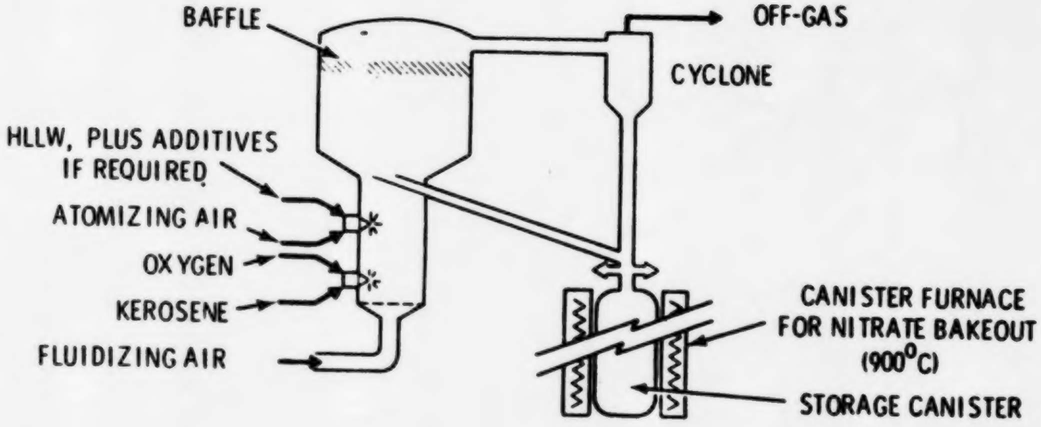

BAFFLE
OFF-GAS
CYCLONE
HLLW, PLUS ADDITIVES IF REQUIRED
ATOMIZING AIR
OXYGEN
KEROSENE
FLUIDIZING AIR
CANISTER FURNACE FOR NITRATE BAKEOUT (900°C)
STORAGE CANISTER

FIGURE B–1 FLUIDIZED-BED CALCINATION

Source: Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle. ERDA 76-43, U.S. Energy Research and Development Administration, May 1976.

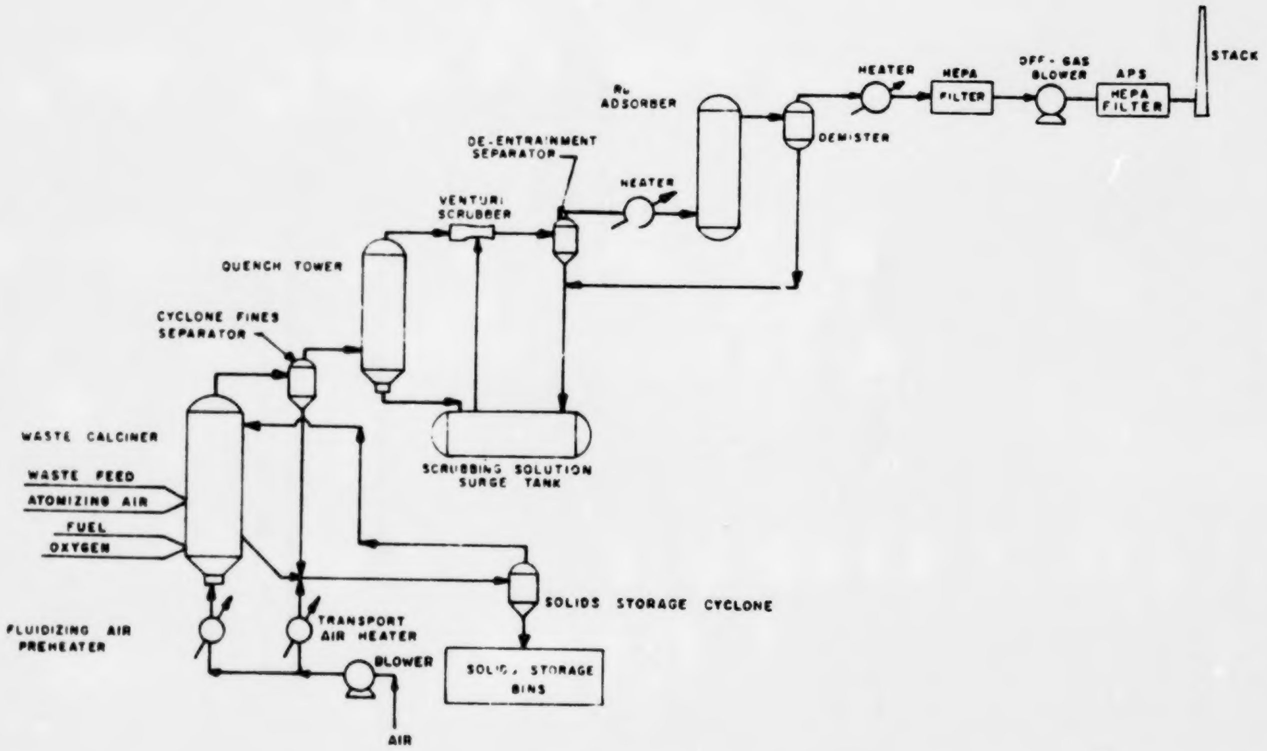

STACK
HEATER
HEPA FILTER
OFF-GAS BLOWER
APS HEPA FILTER
Ru ADSORBER
DEMISTER
DE-ENTRAINMENT SEPARATOR
HEATER
VENTURI SCRUBBER
QUENCH TOWER
CYCLONE FINES SEPARATOR
WASTE CALCINER
WASTE FEED
ATOMIZING AIR
FUEL
OXYGEN
SCRUBBING SOLUTION SURGE TANK
SOLIDS STORAGE CYCLONE
FLUIDIZING AIR PREHEATER
TRANSPORT AIR HEATER
BLOWER
SOLIDS STORAGE BINS
AIR

FIGURE B–2 GENERAL CALCINATION FLOWSHEET FOR THE NWCF

Source: Anderson, F.H., et al. Design Criteria for the New Waste Calcining Facility at the Idaho Chemical Processing Plant. In: Management of Radioactive Wastes from the Nuclear Fuel Cycle. Proceeding Series STI/PUB/433, International Atomic Energy Agency, 1977.

bed calciner solidified relatively low-activity wastes, which were of a substantially different chemical composition than the high-activity fission-product nitrates expected from commercial reprocessing. Pilot-scale testing with simulated (non-radioactive) commercial HLLW feed compositions has been conducted to demonstrate the feasibility of fluidized-bed calcination with commercial wastes. Some difficulty was experienced in the INEL commercial pilot plant studies with agglomeration of sodium at higher than 0.4 M concentration(2) (sodium nitrate exists in a molten, undecomposed state over the temperature range 350-833°C). It is not clear that higher sodium concentrations would necessarily be present in commercial fuel, however, depending on the design of the reprocessing plant. (Sodium is, of course, present in neutralized wastes.) Commercialization tests at INEL showed that addition of powdered iron inhibited agglomeration at 1.13 M sodium. Calcination tests of dilute HLLW revealed some problems in scale-up to commercial-size 30.5-cm (12-in) calciners. The INEL defense waste calcination program experienced plugging of off-gas filters with ruthenium compounds; this did not seem to happen in the commercial program, but the reasons are not clear. The conclusions of the INEL commercialization studies were that fluidized bed calcination could be used for commercial application, but that not all the questions of sodium content, or of off-gas filter plugging, appeared to be fully answered.(7)

The maximum credible accident for a fluidized-bed calcination system has been postulated to be a collapsed bed. If no corrective action were taken, a collapsed bed of one-year-out-of-reactor calcine in a 75-cm diameter vessel would result in a molten core surrounded by sintered calcine. In such a situation, the vessel wall could dissipate the decay heat by convection and radiation at wall temperatures well below the melting point of stainless steel. Nevertheless it appears that the effects of a collapsed bed may be reduced by processing ten-year-old waste and by installing redundant air-supply and bed-discharge systems. The major objectives of the ongoing development program are(2):

1. Verification of the calcinability of simulated Allied General Nuclear Services (AGNS) waste at steady-state conditions in a large-scale pilot plant unit.

2. Verification of the ability of the off-gas cleanup system to remove particulate and volatile fission products and noxious chemicals to acceptable levels.

3. Verification on a suitable scale of the ability to address major safety questions, e.g., removal of a collapsed configuration.

4. Design of a practical calcine storage system.

5. Evaluation of the need for further equipment development.

6. Prototype equipment and remote-maintenance mockup testing as necessary to minimize the risk of significant process or equipment problems during plant operation.

Completion of the development and prototype testing programs is scheduled for 1978. It is expected, based on non-radioactive testing, that all of the data for detailed design would be available in 1977.

B-3.2.1.1.1.2 Continuous Inert-Bed Calcination

If the calcine is to be used for glassmaking, large amounts of silica must be added to it. If, instead, this silica is added to the fluidized-bed calciner, better control of a more stable "inert bed" is achieved, producing a calcine already pre-mixed with silica.

After the INEL studies of commercialization of fluidized-bed calcination, BNWL continued to pursue commercialization studies of fluidized-bed calcination, using an inert-bed process variation. BNWL's process injects an inert material (silica) into the fluidized bed, thereby simplifying calciner operation and allowing higher sodium concentrations in the feed material (up to 1.2 M Na, compared with a limit of 0.4 M Na in the INEL tests). A schematic diagram of such a calciner is shown in Figure B-3.(8)

Agglomeration may be avoided by continuously sweeping the bed with fresh bed material. Since the coated material is continuously withdrawn and the calcine powder is continuously swept from the bed, the concentration of calcine in the bed may be kept low. This reduces the possibility of bed collapse and the need for emergency drains or bed coolers (for the case of loss of fluidizing air).

The inert-bed concept has been shown to be usable over wide operating ranges. Simulated non-radioactive HLLW has been calcined at feed rates up to 40 liters/hr.

BNWL may recommend addition of the glass-forming materials to the liquid waste before it is introduced to the calciner. In this way a single feed inlet would suffice. The one experimental run made using this concept was successful. BNWL is also considering the use of a sand filter instead of sintered metal filters because there is some concern that volatilized materials may plate out on the sintered metal and plug the openings. The sand in a sand filter would be continually changed, with the discharged sand forming the inert bed feed to the calciner.

Design of a scaled-up fluidized-bed calciner has been initiated. Startup and testing is planned for late 1977. Major development objectives include verification of long-term HLLW operation, pilot-scale radioactive testing, and full-scale, remote demonstrations. It is hoped that radioactive runs can be started in 1979. BNWL is quite optimistic about the commercial feasibility of such a process, but prefers the spray calciner for near-term commercialization.

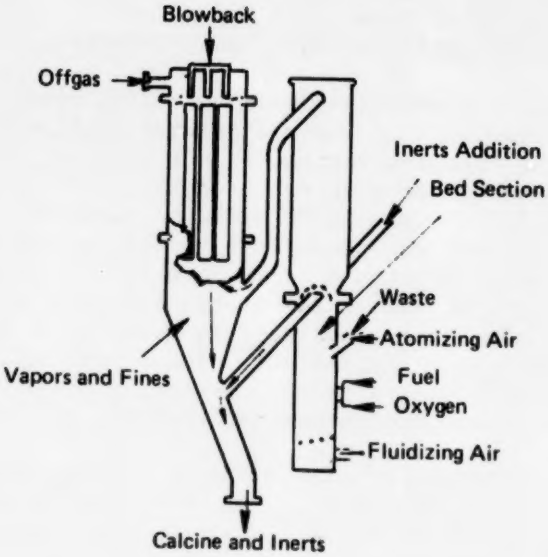

Blowback
Offgas
Inerts Addition
Bed Section
Waste
Atomizing Air
Vapors and Fines
Fuel
Oxygen
Fluidizing Air
Calcine and Inerts

FIGURE B–3 CONTINUOUS INERT BED FLUIDIZED-BED CALCINER

Source: Management of Radioactive Wastes from the Nuclear Fuel Cycle. Proceeding Series STI/PUB/433, International Atomic Energy Agency, 1977.

B-3.2.1.1.1.3 Spray Calcination

Spray calcination does not have as extensive an operating history as fluidized-bed calcination, but it is conceptually simpler and equally compatible with a downstream vitrification system. It has been under development at BNWL for over 15 years. Early spray-calcination programs uncovered difficulties with spray nozzle performance, development of wall scale, and carryover of fines into the off-gas. In current demonstrations these problems appear to have been corrected.

Despite the lack of commercial-scale operating history, spray calcination has been tested in 13 fully-radioactive engineering-scale runs during the Waste Solidification Engineering Prototypes (WSEP) program.(2) According to BNWL, the WSEP program resulted in a total of 600 operating hours and solidified 8700 liters (2300 gallons) of waste. The present BNWL waste fixation program has produced a development spray calciner (44 non-radioactive runs, 875 hours at 70 liters/hr feed) and a full-scale spray calciner (3 non-radioactive runs, 210 liters/hr feed). Actually, the "full-scale" spray calciner is larger than would be required for a 5 MTU/day plant (estimated at 100 liters/hr feed). The spray calciner presently appears to have a significant edge over fluidized-bed calcination as a choice for commercialization.

The full-scale development unit is shown in Figure B-4. An active pilot plant will be placed in operation in 1978. During late 1979, testing of mockup equipment will be used to verify remote design features. Development should be complete by the end of 1980.(5)

The HLLW at about 40°C is pumped to the calciner through a pneumatic spray nozzle. The waste solution spray, about 70 μm diameter droplets, is evaporated and converted into oxides and reaction gases. The final product is very fine (2-5 μm), typically contains less than 0.5% by weight moisture and less than 1% by weight nitrate, and has a density of 0.5-1.3 g/cm^3. The calciner walls are controlled at a temperature of 600-800°C by an external multizone resistance furnace for experimental purposes, but a single-zone heater is expected to suffice for a commercial unit. Deposits on heat transfer surfaces within the calciner are removed by the periodic operation of side-mounted vibrators.

The gases released are primarily H_2O, HNO_3, air and NO_x, with small amounts of CO_x, H_2, O_2, and N_2. The off-gas at about 350°C is passed through sintered stainless steel filters to remove entrained calcine particles. Less than 0.1% by weight of calcine passes through the filters and less than 2% of the ruthenium is estimated to escape to the off-gas system. The balance of the radionuclides, except for volatiles such as iodine, remain in the calcine. The filters are periodically reverse-pulsed with air to prevent the accumulation of an impervious calcine cake. Off-gas is routed to effluent treatment for radionuclide and NO_x removal.

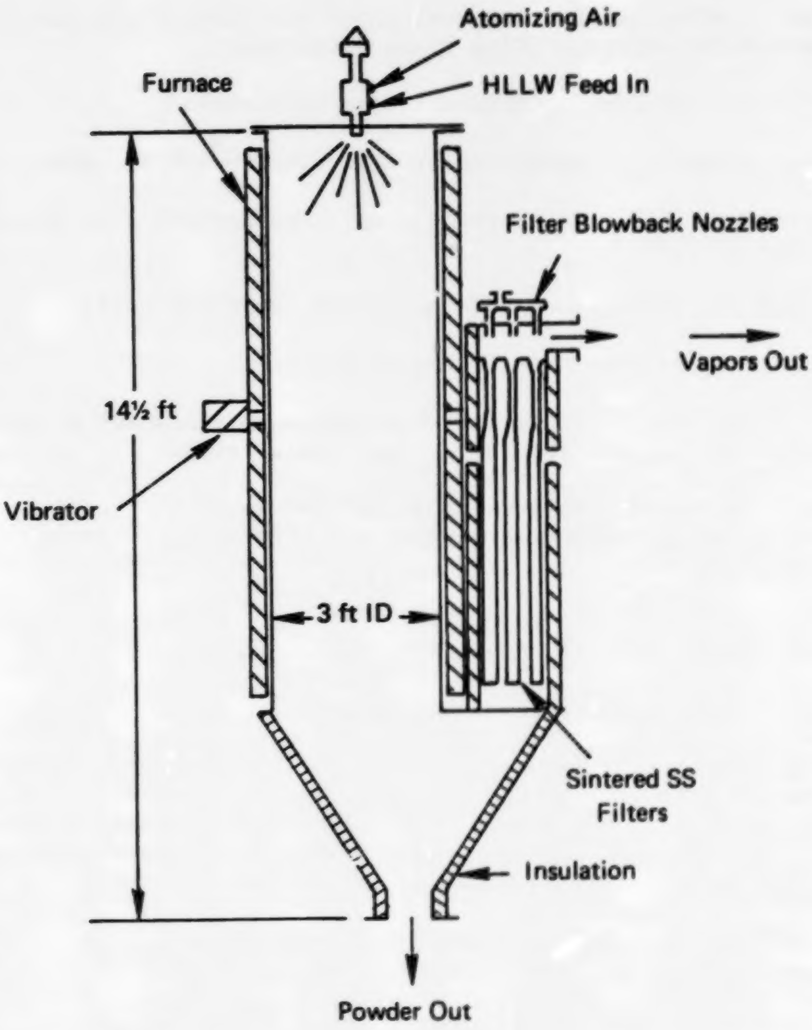

Atomizing Air
Furnace
HLLW Feed In
Filter Blowback Nozzles
Vapors Out
14½ ft
Vibrator
3 ft ID
Sintered SS Filters
Insulation
Powder Out

FIGURE B–4 SPRAY CALCINER FOR 5 MT/DAY REPROCESSING FACILITY

Source: McElroy, J.L. et al. Waste-Solidification Technology U.S.A. In: Proceedings of the International Symposium on the Management of Waste from the LWR Fuel Cycle. Denver, Colo., July 1976. CONF 76-0701.

BNWL favors spray calcination over other calcination processes for near-term commercialization, citing these advantages:

- One moving part (associated with the vibrator);
- Overall simplicity resulting in fast startup and shutdown;
- Calcination of a wide range of waste compositions with sodium content to over 2 M;
- Negligible inventory of calcine entrained in the unit;
- Demonstrated low release of radionuclides;
- Production of a fine calcine well-suited to glassmaking (but perhaps not as desirable as a final waste form).

The major development objectives if calcine is to be the final product would be to increase bulk density and thermal conductivity of the calcine product.(2)

B-3.2.1.1.1.4 Rotary Kiln Calcination

The French program to solidify HLLW dates from 1959. The early work was centered mostly on a pot vitrification process (called "PIVER"). Since batch processes have limited capacity, it was decided to develop a continuous rotary kiln calciner coupled to a continuous melter. After 5000 hours of testing of the rotary kiln calcination process, a full-size industrial rotary kiln was manufactured in 1971. Testing on this unit started in 1972 and is still continuing.(2)

Figure B-5 is a schematic diagram of a rotary kiln calciner. It consists of an externally-heated (500°C) rotating cylinder about 3.5 m long by 27 cm diameter, operating at a slight slope. Deacidified HLLW is dried and almost completely denitrated before it is discharged. A loose bar within the kiln keeps the calcine free-flowing and prevents it from sticking to the wall. The calcine product is further heat-treated in a canister furnace at 900°C to assure total decomposition of the nitrate.

Air pressure in the kiln is maintained slightly negative relative to the ambient pressure. The kiln is heated externally by a four-zone electric resistance furnace. Average processing time for conversion of HLLW to calcine is about 4 minutes.(4)

Because the French have always fed the calcine directly to a vitrification unit, no actual data on the calcine are available. It has a bulk density estimated at 1.0-1.3 g/cm^3.

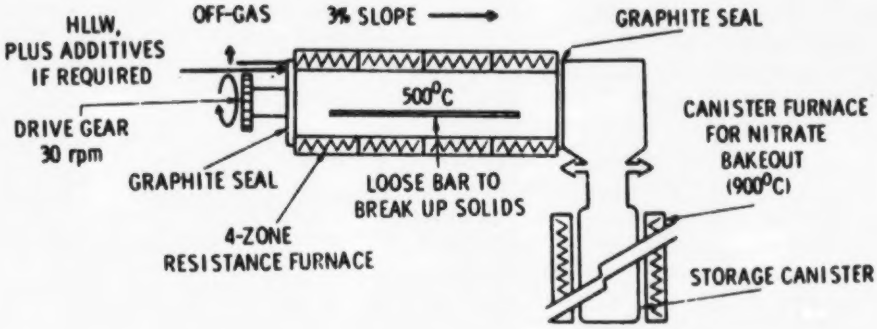

HLLW, PLUS ADDITIVES IF REQUIRED
OFF-GAS
3% SLOPE
GRAPHITE SEAL
500°C
DRIVE GEAR 30 rpm
CANISTER FURNACE FOR NITRATE BAKEOUT (900°C)
GRAPHITE SEAL
LOOSE BAR TO BREAK UP SOLIDS
4-ZONE RESISTANCE FURNACE
STORAGE CANISTER

FIGURE B–5 ROTARY KILN CALCINATION

Source: Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle. ERDA 76-43, U.S. Energy Research and Development Administration, May 1976.

This process has been under development in France for more than ten years in connection with a continuous vitrification process. During much of this time an engineering-scale unit has been in use with simulated non-radioactive waste. A radioactive engineering-scale rotary calciner is scheduled to be in operation in 1977 as part of a vitrification facility. Much of the technology developed from this installation would be applicable to a calcine product facility, were it desired to stop at the calcine state. In that case, the major development objectives for a facility with calcine as a final product would be to increase the bulk density and the thermal conductivity of the calcine.(2)

B-3.2.1.1.2 Glassification

Borosilicate glass is the preferred form in most glass programs (U.S., U.K., France, India, and Germany). It has relatively low temperatures of formation, low corrosiveness to container material during formation, and low leach rates. Borosilicate glass typically has the properties shown in Table B-7.

The manufacture of glass in a radioactive facility is complicated by the difficulties of remotely operating and maintaining equipment; special attention must be given to questions of reliability. In this regard, simple, rugged equipment may have substantial advantages over more complex (and more sensitive) equipment. In addition, since the long-term stability of the radioactive glass product cannot be measured directly, special problems arise in the design and operation of process control instrumentation in order to insure uniform product quality.

B-3.2.1.1.2.1 In-Can Melting

In the in-can melting process, the calcine produced from HLLW is dropped into a storage canister along with frit material, and is melted in the storage canister, using a multizone furnace below the calciner. Off-gas from the melting mixture is vented to the calciner off-gas system. Because heat is generated by the waste, the temperature below the melt level would tend to rise as the canister fills; the lower zones of the furnace, therefore, are turned off as the melt level rises above them and cooling is initiated. After the canister is filled, the calcine and frit are diverted to another canister in a parallel furnace.

A schematic diagram of the in-can melter coupled to a spray calciner is shown in Figure B-6.

After cooling, the canister is capped and removed from the furnace. The cap is seal-welded and the canister is checked for leaks, and decontaminated prior to being sent to storage.

TABLE B-7

CHARACTERISTICS OF BOROSILICATE GLASS

Composition	
SiO_2	25-40 wt%
B_2O_3	10-15 wt%
Alkali Metal Oxides	5-10 wt%
ZnO	0-20 wt%
Waste Oxides	20-35 wt%
Density	3.0-3.6 g/cm^3
Thermal Conductivity	0.9-1.3 W/(m-°C)
Processing Temperature	1000-1400°C

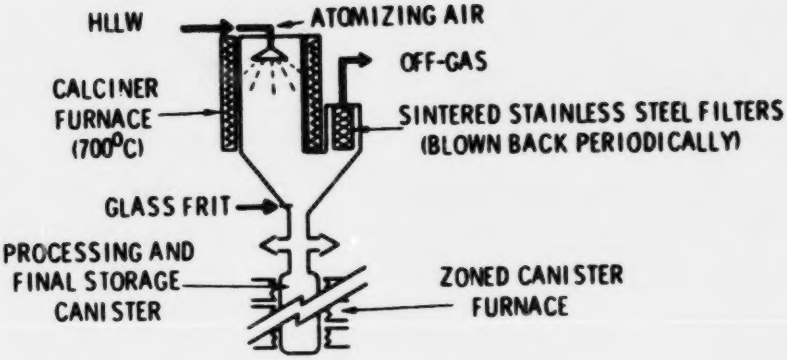

HLLW
ATOMIZING AIR
OFF-GAS
CALCINER FURNACE (700°C)
SINTERED STAINLESS STEEL FILTERS (BLOWN BACK PERIODICALLY)
GLASS FRIT
PROCESSING AND FINAL STORAGE CANISTER
ZONED CANISTER FURNACE

FIGURE B–6 IN-CAN MELTING

Source: Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle. ERDA 76-43, U.S. Energy Research and Development Administration, May 1976.

The in-can melting product monolith is described as follows: "Because the melting is done in a metallic storage canister, processing temperature restraints prevent complete assimilation of all waste oxides into the glass matrix. The result is a glass matrix in which small amounts of several crystalline phases are dispersed. Cerium dioxide is the most prevalent crystalline phase present. The leachability of the glass is not significantly increased by the crystalline phases."(2)

On the formerly prevalent assumption that wastes might be relatively fresh, several techniques have been tried to increase the heat removal rate within the glass. Installation of internal longitudinal fins in the canister increases thermal conduction, increases processing capacity, and approximately doubles the amount of heat-producing radioisotopes allowable in the canister.

According to BNWL, the advantages of the in-can melting process are that it(2):

- Minimizes process steps and equipment;
- Does not require transfer of melt;
- Assures that everything entering the melter, with the exception of some volatile species, is fixed in the storage canister;
- Eliminates the problem of melter deterioration and disposal;
- Is not affected by the addition of reducing agents for phase separation control; and
- Has been demonstrated both in the United States and abroad.

BNWL has conducted radioactive tests of in-can melting at 7.5 kg glass/hr.(2) The on-going waste fixation program at BNWL has produced 2500 kg of melt in 22 engineering-scale runs (non-radioactive) at melting rates up to 50 kg/hr in 30-cm (12-in) diameter cans.

To insure continuous smooth operation of the in-can melt, the process requires (1) temperature controls (zoned-furnace and controlled-heating/cooling requirements complicated by canister sensitization temperature limits and internal heat generation), and (2) level instrumentation (level is an important input to the temperature controls). There nonetheless seems to be little doubt as to the commercial practicability of in-can melting as an option, and the product is well defined.

Major development objectives in the current in-can melting program include final specification of canister design; optimization of process procedures, using a multizone furnace; and full-scale remote demonstrations.(2)

B-3.2.1.1.2.2 Continuous Ceramic Melter

This process (also called "joule heating"), may ultimately prove superior to in-can melting; it has been commercially utilized (for 30 years) in the glass industry. It is carried out in a ceramic-lined melter with internal electrodes. In such a unit, molten glass acts as its own electrical resistance heating element. This type of melter is popular in the glass industry because of its high capacity and long life while producing glass of superior quality with low off-gas effluents.(2) These same characteristics are desirable for an HLLW vitrification system. The process has been developed at BNWL; the French and the Danes have also explored joule heating melters. The work at BNWL started in 1973 and is described in ERDA 76-43.(2) Following laboratory-scale tests in 1974, an engineering prototype was built and kept at 1000-1300°C continuously for 11 months. Intermittent operation during this period produced a total of 4 MT of glass at rates up to 60 kg/hr. During this time only minor corrosion of the refractories and electrodes was noted. The unit was restarted in January 1976 and has accumulated over 23 months of exposure to molten glass without failure (including the time prior to restart).

Figure B-7 is a schematic diagram of a joule-heated continuous ceramic-line melter. Sufficient heat can be transferred to allow drying of the waste as well as melting, possibly eliminating the need for a separate calciner. In this case, liquid waste would be fed directly to the melter.

To start operation of such a unit, the glass must be brought to a temperature at which it becomes adequately conductive. Sacrificial heating elements were initially used, but once the glass became molten, the sacrificial element was dissolved by the glass. BNWL has had success with a recently-developed startup/restart technique in which sodium hydroxide solution is used as the initial carrier of the electric current. This eliminates the need for temporary electrodes and resistance elements. For initial startup, the melter is charged with a 20-cm (8-in) depth of glass frit. NaOH solution is then added to conduct the electricity until the glass starts to melt. The NaOH also acts as a flux, which causes the glass to melt at a lower temperature.

Once startup is complete, calcine and frit may be added as molten glass is drawn off. With calcined waste and frit fed to the melter, capacities higher than 20 kg of glass/hr/100 cm^2 of surface area have been demonstrated.(2) The BNWL melter has been operated at rates as high as 85 liters/hr.

A tilt-pour mechanism has been devised to eliminate the need for a freeze valve (a potential source of failure).

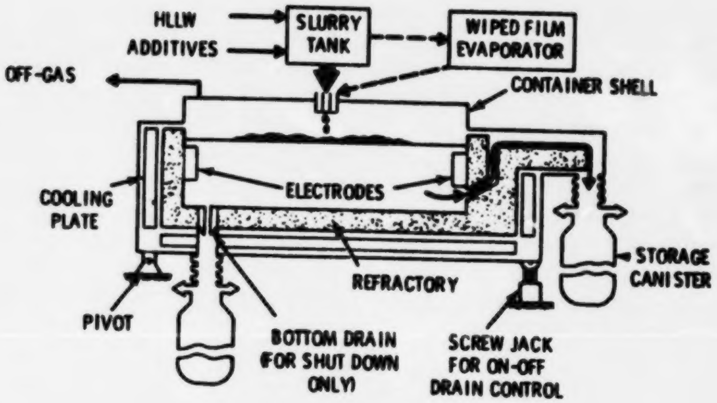

HLLW
ADDITIVES
SLURRY TANK
WIPED FILM EVAPORATOR
OFF-GAS
CONTAINER SHELL
ELECTRODES
COOLING PLATE
STORAGE CANISTER
REFRACTORY
PIVOT
BOTTOM DRAIN (FOR SHUT DOWN ONLY)
SCREW JACK FOR ON-OFF DRAIN CONTROL

FIGURE B–7 PROCESS SUMMARY – CONTINUOUS CERAMIC MELTER

Source: Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle. ERDA 76-43, U.S. Energy Research and Development Administration, May 1976.

Some of the advantages of the ceramic melter are:

- Long (demonstrated) melter life,
- Flexibility in waste input composition,
- High capacity per hot-cell area,
- Ability to produce molten glass for monoliths, marbles, coated pellets, etc.

The engineering-scale melter has been shown capable of recovering from electric power outages (more than 25 min), sudden additions of liquid solution, periodic under- and over-powering, and the addition of metal.(9) The process must now be demonstrated with extended remote operation using radioactive wastes for feed.

A more advanced version of this process involves direct liquid feeding of the HLLW to the ceramic melter. This has been tested at BNWL, and a capacity of more than 6 liters/hr of HLLW/1000 cm^2 surface area has been demonstrated.(2) If direct liquid feeding is employed, the waste is transferred to a mix tank, mixed with frit, and the slurried waste is then fed by gravity into the melting cavity; there, heat from the glass pool evaporates the liquid to steam (the safety of this step will have to be demonstrated) and decomposes the metal nitrates to oxides that dissolve in the glass. Even if the molten surface is completely covered with 20-40 mm of solution, no loss in processing rate is noted and less than 0.5% of the waste is entrained in the off-gas stream. This approach is also being pursued at BNWL.

B-3.2.1.1.2.3 French Rotary Kiln - Continuous Metallic or Ceramic Melter

The French rotary kiln calciner described under Section B-3.2.1.1.1.4 has always been closely coupled to a vitrification furnace. Figure B-8 is a schematic diagram of the kiln coupled to an induction heater to convert the calcine into a borosilicate glass.

The Inconel, induction-heated, vitrification furnace is connected directly to the rotary kiln calciner by an expansion bellows. Glass frit and calcine from the kiln are continuously added to the furnace (1 m high x 35 cm diameter), where they are melted.

The level of molten glass in the furnace is allowed to rise to a preset value, and the glass then flows through a freeze valve into a product container. Two vitrification units, each of 20 kg/hr product capacity, are connected to each kiln.(2) The off-gas from the vitrification units is routed through the rotary kiln and becomes part of the kiln off-gas.

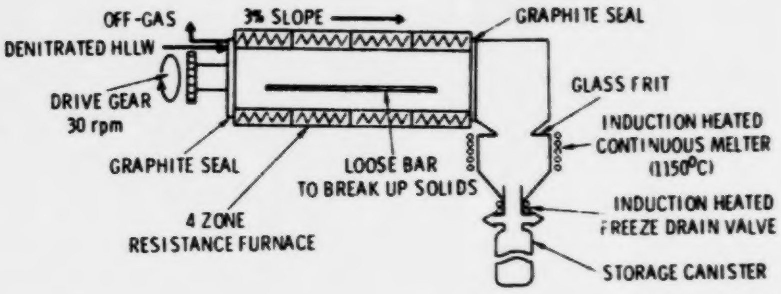

OFF-GAS
3% SLOPE
GRAPHITE SEAL
DENITRATED HLLW
DRIVE GEAR
30 rpm
GLASS FRIT
INDUCTION HEATED
CONTINUOUS MELTER
(1150°C)
GRAPHITE SEAL
LOOSE BAR
TO BREAK UP SOLIDS
INDUCTION HEATED
FREEZE DRAIN VALVE
4 ZONE
RESISTANCE FURNACE
STORAGE CANISTER

FIGURE B–8 FRENCH ROTARY KILN – CONTINUOUS METALLIC MELTER

Source: Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle. ERDA 76-43, U.S. Energy Research and Development Administration, May 1976.

The final disposal canisters are exposed to somewhat lower temperatures than with in-can melting, but there are also the disadvantages of potential rapid melter deterioration (discovered during the PIVER program, when vessel replacement was required every 30 cycles), and the need for a freeze valve (malfunction or plugging of which shuts down the entire operation). Nonetheless, the French had sufficient success with this arrangement to incorporate a melter vessel into the new "AVM" facility at Marcoule, which will be capable of handling the entire HLLW output from the reprocessing plant.

The major development objective for the near future is to test the system under high-level radioactive conditions.

B-3.2.1.1.2.4 German Spray Calciner-Continuous Melter

Research and development on the glassification of radioactive wastes has been under way in Germany for about ten years. The "VERA" process being developed in Karlsruhe is the most advanced of the German programs. Figure B-9 is a schematic diagram of the original basic equipment configuration.

Initially the waste solution is denitrated by reaction with formic acid. (The volatilization of ruthenium is thus reduced to 0.01% of the total ruthenium content.) The waste is then pumped to the top center of the spray calciner where it is steam-atomized, then dried and calcined in flight by super-heated (650°C) steam. The calcine is removed by 3-micron sintered metal filters, and is blown back periodically to fall into the melter. Addition of SiO_2 to the HLLW seems to reduce filter clogging.

In the induction-heated Inconel melter, the calcine and the glass frit are heated to about 1150°C for about two hours. Molten glass is drained from the melter via a freeze valve into the final storage canister (stainless steel).

A later concept combined the denitrator, calciner, and filter into a single integrated unit, reducing the size of the unit itself and of the hot cell.(10) This combination unit has successfully processed at least 3000 liters of simulated HLLW so far. A flowsheet is shown in Figure B-10. Complete condensation of the heating steam and recycling of the condensate decreases the radioactivity of the off-gases by several orders of magnitude.(10)

A still later variation of the VERA process consists of a modified calciner and ceramic melter integrated into a single unit. The simulated high-level waste, mixed with glass frit, is sprayed directly into the wall-heated calciner/melting unit where the droplets are dried and the product is melted to borosilicate glass at 1200°C. In this version the heating steam circuit is eliminated. A large portion of calcining heat is supplied by radiation from the ceramic melter directly below the calciner.

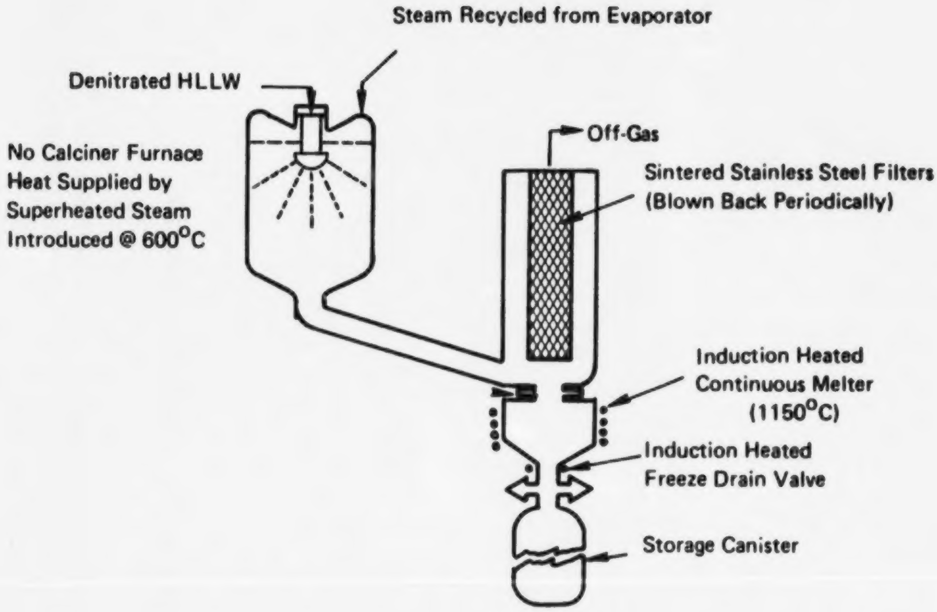

Steam Recycled from Evaporator
Denitrated HLLW
No Calciner Furnace
Heat Supplied by
Superheated Steam
Introduced @ 600°C
Off-Gas
Sintered Stainless Steel Filters
(Blown Back Periodically)
Induction Heated
Continuous Melter
(1150°C)
Induction Heated
Freeze Drain Valve
Storage Canister

FIGURE B–9 ORIGINAL VERA PROCESS

Source: Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle. ERDA 76-43, U.S. Energy Research and Development Administration, May 1976.

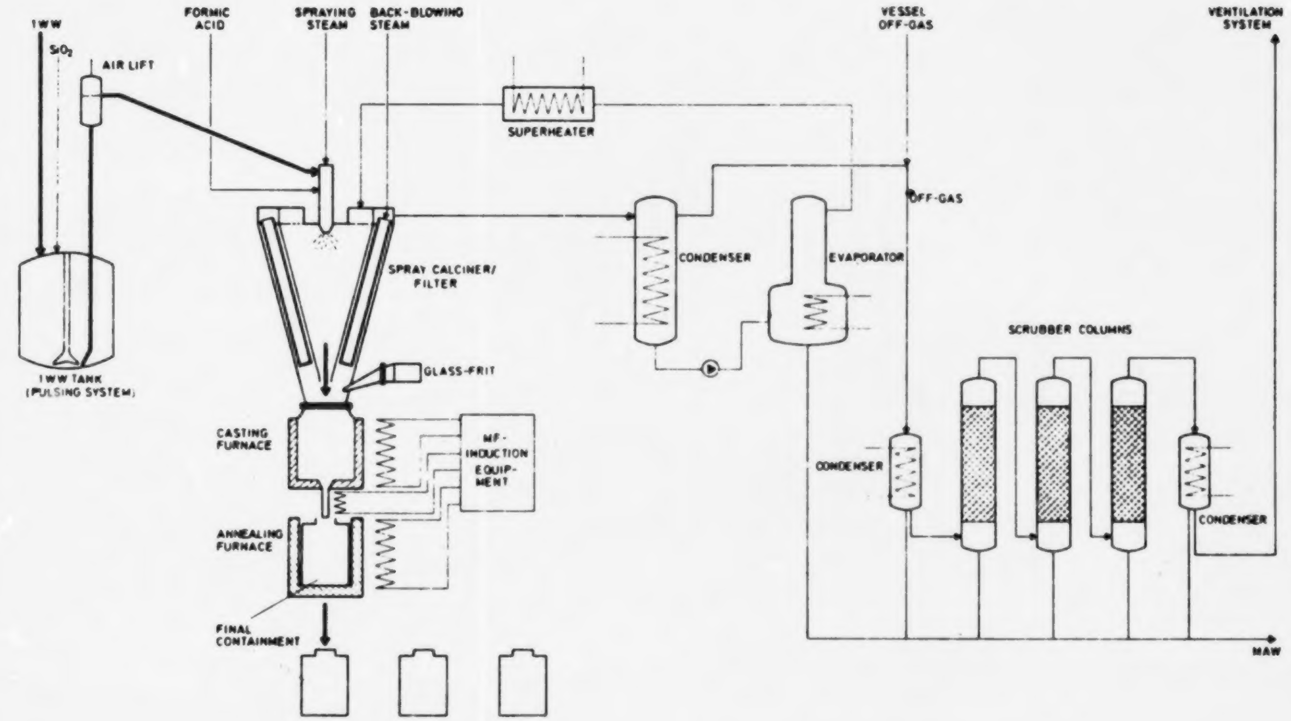

1WW
SiO_2
AIR LIFT
FORMIC ACID
SPRAYING STEAM
BACK-BLOWING STEAM
SUPERHEATER
VESSEL OFF-GAS
OFF-GAS
VENTILATION SYSTEM
CONDENSER
EVAPORATOR
SPRAY CALCINER/ FILTER
SCRUBBER COLUMNS
1WW TANK (PULSING SYSTEM)
GLASS-FRIT
CASTING FURNACE
MF-INDUCTION EQUIPMENT
CONDENSER
CONDENSER
ANNEALING FURNACE
FINAL CONTAINMENT
MAW

FIGURE B–10 VERA PROCESS SYSTEMS

Source: Management of Radioactive Wastes from the Nuclear Fuel Cycle. Proceeding Series STI/PUB/433, International Atomic Energy Agency, 1977.

The pilot-scale melter with a design capacity of 30 liters of waste solution per hour started operation in September 1976. In the first long-range test of 100 hours a total of 6600 liters of simulated high-level waste solution was converted to 1500 kg of glass that was filled into 25 steel cylinders.(5)

Work with the ceramic melter is expected to continue, culminating in a radioactive prototype installation in 1983 or 1984.(11)

Compared with the U.S. spray calciner design, the German VERA unit has the advantage of elimination of HNO_3 and reduction of volatile radioactivity in the off-gas by injection of formic acid. On the other hand, the German design requires a more complicated atomizing nozzle design, which increases potential for clogging from waste dried by the steam heat before passing through the nozzle opening.

B-3.2.1.1.2.5 British Rising Level Glass Process (FINGAL/HARVEST)

Development of a rising level vitrification process called FINGAL was begun in the late 1950's at Harwell, England.(5) This process fed HLLW, together with an aqueous suspension/solution of silica and sodium tetraborate, into a stainless steel cylinder at 1050°C in a multizone resistance heated furnace (see Figure B-11). Evaporation, denitration, sintering, and glass formation proceeded simultaneously during the filling cycle. The feed rate was kept constant and the free liquid level rose at a rate equal to the rate at which glass was produced. The off-gases from the first, or glassmaking, vessel were passed through the second and third vessels, which contained filters to trap particulate material and volatile ruthenium. When the first vessel was filled with glass, it was removed to storage and the vessels in the other positions were progressively moved into the furnace. The filter in each cylinder was incorporated into the glass melt as vitrification proceeded in the furnace.

The FINGAL process was shut down in early 1966 after completing 72 runs, of which 8 involved processing radioactive wastes from the Windscale works. In 1972, it was decided to resume development of a solidification process that would be suitable for full-scale industrial use. The modified process is called HARVEST.

The HARVEST concept differs from the FINGAL program in batch size, off-gas system, and geometry of the container vessel. The maximum batch size for the FINGAL plant is about 80 kg of glass; the HARVEST batch size is intended to be 1-3 MT. A HARVEST pilot plant has been constructed at Harwell. Figure B-12 is a flow diagram of the new pilot plant.

The main advantages of the rising level glass process are the elimination of a separate calciner and use of the process vessel as the final storage canister.

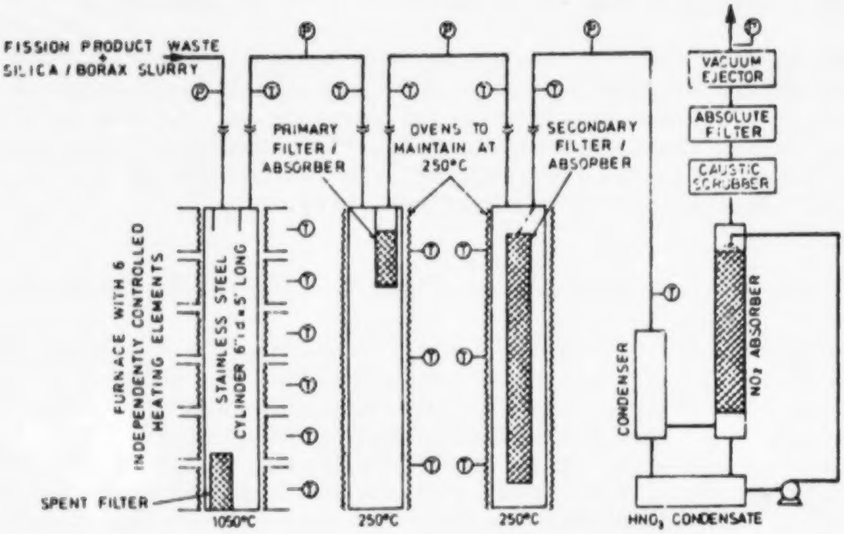

FISSION PRODUCT WASTE
+
SILICA / BORAX SLURRY
P
T
PRIMARY FILTER / ABSORBER
OVENS TO MAINTAIN AT 250°C
SECONDARY FILTER / ABSORBER
VACUUM EJECTOR
ABSOLUTE FILTER
CAUSTIC SCRUBBER
FURNACE WITH 6 INDEPENDENTLY CONTROLLED HEATING ELEMENTS
STAINLESS STEEL CYLINDER 6"i.d.×5' LONG
SPENT FILTER
1050°C
250°C
250°C
CONDENSER
NO₂ ABSORBER
HNO₃ CONDENSATE

FIGURE B–11 SIMPLIFIED FLOWSHEET OF FINGAL PROCESS

Source: Techniques for the Solidification of High-Level Wastes. Technical Report Series 176, International Atomic Energy Agency, 1977.

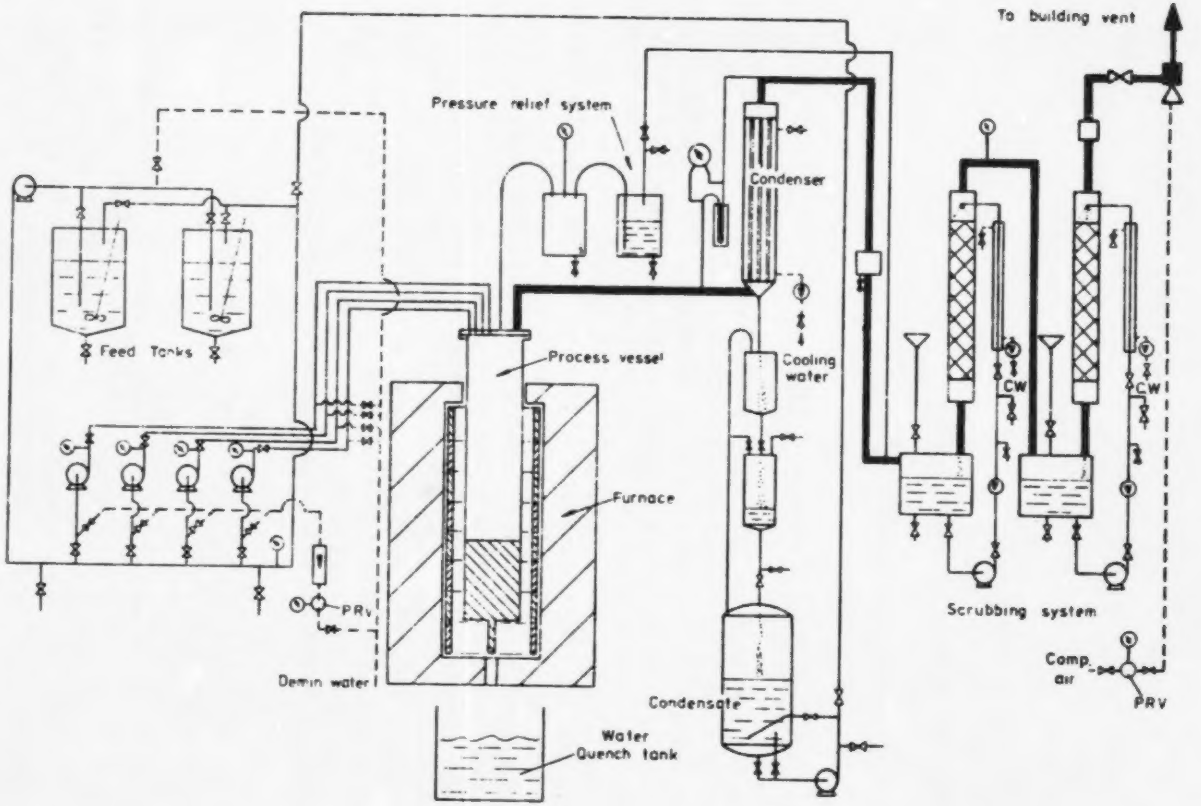

To building vent
Pressure relief system
Condenser
Feed Tanks
Process vessel
Cooling water
CW
CW
Furnace
PRV
Demin water
Scrubbing system
Comp. air
PRV
Condensate
Water
Quench tank

FIGURE B–12 NEW HARVEST PILOT PLANT FLOW DIAGRAM

Source: Management of Radioactive Wastes from the Nuclear Fuel Cycle. Proceeding Series STI/OUB/433, International Atomic Energy Agency, 1976.

B-3.2.1.2 Solidification Processes in Early Stages of Development

Several processes under development were considered too preliminary for consideration at this time. They are:

1. Supercalcine
2. Sintering
3. Metal Matrices
4. Glass-ceramics
5. Coated Pellets
6. Ion Exchange
7. German Thermite Process
8. German (Julich) Borosilicate Glass Process
9. German (Pamela) Phosphate Glass Process

These processes are discussed in Appendix B-II.

B-3.2.2 Cladding Hulls and Fuel Bundle Residues

Current plans for commercial fuel reprocessing include an acid leach of fuel bundles which have first been sheared into short lengths. The solid residue from the leach consists of fuel cladding, fuel bundle support rods, poison rods, end fittings, fuel support grids, springs, and spacers. This hardware weighs about 325 kg/MTU. As recovered from the acid dissolver, the hardware residues are a relatively low-density (about 1 kg/liter) waste. The composition of these residues will vary as fuel manufacturers change their materials of construction. The current composition is mainly Zircaloy, with lesser amounts of stainless steel, Inconel, and other materials. The cladding, after being acid-leached, contains about 0.1% of the actinide content of the original fuel; fission product content of the cladding is assumed to be higher (0.2%) to allow for the effect of fission fragment recoil into the cladding. For actinides, ERDA 76-43(2) gives an estimated range of 0.05-0.5%; the AGNS Final Safety Analysis Report (FSAR)(12) and ORNL(13) settle on 0.1% as the most likely value. NFS experience was that 0.05% of the fuel activity was contained in the hulls.(14)

Table B-8 gives expected properties of a fuel bundle waste stream for an LWR fuel (assumed to be PWR fuel).

If the cladding is assumed to be compacted to 70% of its theoretical density(2) the final volume would be about 60 liters/MTU.(13) This compares with 60-80 liters/MTU of waste glass produced from the HLLW.(2) Thus, the volume of hulls and fuel bundle residues would, if thoroughly compacted, approximately equal the volume of high-level glass from the same fuel.

TABLE B-8

FUEL BUNDLE PROPERTIES

Fuel Assembly Parameters

Total assembly weight	666.8 kg
Zr-4 fuel cladding/assembly	110.5 kg
Zr-4 end plugs/assembly	3.4 kg
Zr-4 in control rod and instrument tubes/assembly	17.9 kg
Total Zr-4/assembly	131.8 kg
Total effective Zr-4/assembly(*)	122.8 kg
Total Inconel 718/assembly	6.2 kg
Total 304SS/assembly	5.3 kg
Total effective 304SS/assembly(*)	2.0 kg
Fuel weight/assembly, heavy metal oxide	523.5 kg
Fuel weight/assembly, heavy metal content	461.4 kg
Weight of total assembly structure(+)/ weight of fuel, kg structure/kg heavy metal	0.3107
Overall assembly length	4.1 m
Overall assembly length with rod cluster control assembly in place	4.3 m
Fuel rod length	3.9 m
Active fuel length	3.7 m
Assembly cross section	.21 x .21 m
Specific power	38.4 kW/kgHM

---

*Weights corrected so that effective weight reflects the average axial flux. This is the value used in the ORIGEN analysis for structure activation. Correction based upon Westinghouse data for the axial flux profile for a 3.7-m active core.

+All non-fuel assembly components.

The expected radioactivity and heat generation of accumulated cladding hulls has been discussed in the Task A report. Total volume through the year 2000 is on the order of 5000 m^3.

The only commercial disposal of cladding hulls and fuel bundle residues to date has been done by Nuclear Fuels Services, Inc. (NFS). In this case, the fuel assembly nozzles or end boxes were removed from the fuel bundle prior to chopping. These end boxes, containing no appreciable uranium or plutonium, were packed into drums separately. The cladding wastes, including hulls cut into short sections, fines, and grids were processed through the leaching cycle, monitored for undissolved fuel, and dumped into 114-liter (30-gallon) steel scrap drums. The drums of hulls and the drums of nozzles or end boxes were placed in burial holes at an on-site facility designated for this purpose, and backfilled with at least 1.3 m (4 ft) of earth cover over the top drum. NFS observed clad waste quantities (uncompacted) of 350 liters/MTU.(14)

The AGNS FSAR indicates much the same procedure, except that it was not planned to separate the end fittings from the hulls before leaching.(12) After leaching, the dissolver baskets were to have been monitored and then the hulls dumped into a hull disposal container. Since Zircaloy fines can be pyrophoric under certain conditions, provision was to have been made to add sand to the disposal container if the hulls should ignite. The container was to have a capacity of 1470 liters (0.9 m i.d. x 2.24 m high) which is sufficient for the waste components from 3 MTU (3 dissolver baskets), equating to about 490 liters/MTU. The AGNS container shell was to have 10-cm (4-in) thick sidewalls and a 15-cm (6-in) thick bottom of integrally cast reinforced concrete with anchor bolts cast into the sides for securing the cover, fabricated from 3.2-cm (1.25-in) thick carbon steel. A rubber-covered asbestos gasket was to provide the seal between the cover and container. The exterior surface of the container was to have been covered with an epoxy coating to aid in decontamination with a high-pressure hose. The hull container was to have been placed in a 1.52-m (5-ft) diameter, 4-m (13-ft) deep hole in the ground. The hole was to have been backfilled and compacted in order to leave a minimum earth cover of 1.22 m (4 ft) above the top of the cask. The area over the buried casks was to have been mounded to shed rain water.

Despite these plans for surface burial, it now appears that because of their TRU contamination (see Task A report), cladding wastes will probably be handled by geologic disposal in a manner similar to that of high-level wastes. At 0.1% of fuel plutonium and fission product content, fresh cladding wastes are contaminated with approximately 3 kg/m^3 actinides, activity levels of 2×10^5 Ci/m^3, and heat rates of approximately 1 kW/m^3.(13) It will be shown in Section B-4.7 that costs for disposal of low-level TRU wastes in deep geologic formations will exceed \$860/m^3. Cladding disposal costs should be above this figure, a substantial economic incentive for volume reduction, and a number of possible treatment steps may be warranted or required.

Figure B-13 shows various possibilities for chop-leach fuel bundle residue treatment. There are many alternative choices among such variables as mixing agents, melting techniques, and conversion processes. Preliminary sorting of residues for size and material, or surface cleaning for decontamination and oxide removal further extends the treatment possibilities and the range of waste product forms. Except for segregation and the dissolving of Zircaloy in aqueous fluorides, all the pretreatment and treatment technologies are in the small pilot plant or conceptual stages requiring further development.(2)

The following discussions of cladding-waste technologies are adapted from ERDA 76-43.(2)

B-3.2.2.1 Pretreatment

Sorting (material segregation) and surface cleaning are the principal pretreatment options. Sorting fuel cladding by size or material may be desirable for reasons of safety, simplified processing, and storage. Sorting can be done before or after the chop-leach process. The recovery of end fittings, grids, guide tubes, etc., as non-TRU wastes and improved efficiency of separation by materials are incentives for fuel bundle disassembly and segregation prior to the shearing operation. Because the fuel assembly parts that have high TRU levels (hulls) are readily distinguishable mechanically from other parts, this step should be reasonably inexpensive.

If an overall evaluation of safety and economics indicates a need to sort the cladding wastes (as appears likely at probable disposal costs of thousands of dollars/m^3), then three size classifications seem desirable. The massive group would contain end fittings and grid sections; the intermediate group, the hulls and plenum springs; and the third group, the remaining material. Undissolved fuel would be in the intermediate or fines group. The fines could be rendered safe for handling by casting in a matrix or by conversion to a more stable compound through controlled oxidation or chemical change.

The design of the General Electric (GE) fuel processing plant at Morris, Illinois included provisions for segregation of fuel bundle residues. Equipment was installed to withdraw fuel rods from the bundle, prior to running the fuel rods through a shear. After leaching, the hulls were rinsed to remove fines. This operation was demonstrated on cold fuel bundles, but was not used on irradiated materials.

Surface treatment is another pretreatment option with possible advantages. It can either reduce the radioactivity of the major volume stream or enhance the characteristics of the final waste form. Zircaloy fuel cladding contains as a residue about 0.1% of the original fuel. About 50% of this is leachable and 50% is nonleachable and integral with the corrosion product oxide on the fuel surface clad.(2) Usually

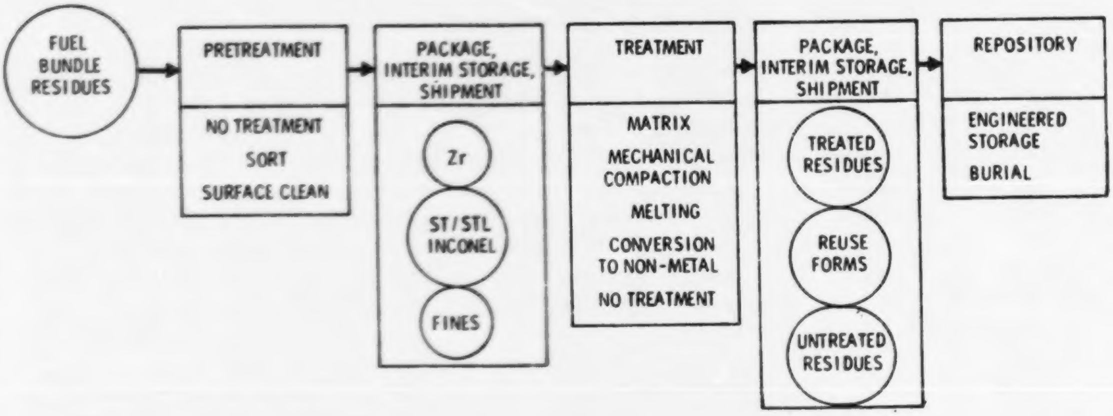

FUEL BUNDLE RESIDUES
PRETREATMENT
NO TREATMENT
SORT
SURFACE CLEAN
PACKAGE, INTERIM STORAGE, SHIPMENT
Zr
ST/STL INCONEL
FINES
TREATMENT
MATRIX
MECHANICAL COMPACTION
MELTING
CONVERSION TO NON-METAL
NO TREATMENT
PACKAGE, INTERIM STORAGE, SHIPMENT
TREATED RESIDUES
REUSE FORMS
UNTREATED RESIDUES
REPOSITORY
ENGINEERED STORAGE
BURIAL

FIGURE B–13 DECISION AREAS IN THE MANAGEMENT OF FUEL BUNDLE RESIDUES

Source: Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle. ERDA 76-43, U.S. Energy Research and Development Administration, May 1976.

less than 0.1% of the TRU element contamination of the cladding is found in the metal. Thus, surface decontamination factors (DF) of about 10^3 are attainable by cleaning the zirconium dioxide product from the surface. Whether it would prove economically advantageous to provide surface cleaning, even at these levels of decontamination, is dependent upon a number of variables, such as regulatory differentiation between higher and lower orders of TRU contamination, and the extent to which the decontamination wastes may be reduced in volume and stabilized. Even a DF of 10^3 does not appear attractive economically if the proposed 10 nCi/g rule is left unmodified, for a typical hull waste contamination level of 10^6 nCi/g would still have to be disposed of in geologic formations after a contamination reduction of 10^3. However, if the decontamination residue can be made considerably more stable than the surface contamination (as seems likely), then a technical case could certainly be made for decontamination. In summary, a conflict may exist between the direction in which the current form of proposed regulations push hull disposal and implementation of the best technology for treating those hulls.

B-3.2.2.2 Treatments

B-3.2.2.2.1 Matrix Formation

Incorporation of the hull waste into a matrix can reduce pyrophoricity, leachability, and requirements for shielding. Even if fines are separated at the reprocessing plant, a matrix binder will reduce the formation of fines in subsequent handling operations. Some form of precompaction or flattening would probably be desirable prior to matrix casting to increase the hulls-to-matrix ratio. Densification and compaction are discussed briefly later in this report. The matrix technology is essentially available.(2)

B-3.2.2.2.1.1 Concrete Matrix

Concrete is an obvious matrix material for immobilizing cladding hulls, based on limited experience with concrete matrices for low- and intermediate-level wastes.(2) There has been concern regarding potential maintenance problems at a disposal facility for concrete-matrix hulls, but the concept appears sufficiently simple to allow for dealing with these concerns.

B-3.2.2.2.1.2 Bitumen Matrix

Bitumen has been used in a radioactive waste disposal system for handling low- and intermediate-level wastes, but it does not appear to be suitable for highly radioactive solid metal waste materials because of radiolytic decomposition of the bitumen.(2) The potential decomposition problem, together with flammability, has discouraged serious consideration of the use of a bituminous matrix for high-level wastes.

B-3.2.2.2.1.3 Glass Matrix

It is conceivable that HLW glasses could be used as a matrix material for hulls. In so doing, two waste forms could be combined, resulting in volume and weight savings. Interactions between glass and zirconium metal alloys would have to be investigated to determine the effects of the zirconium on the glass properties.

B-3.2.2.2.1.4 Sand Matrix

AGNS incorporated provision for adding sand to the hulls before burial, but intended to do so only if the hulls ignited due to pyrophoricity. NFS, before abandoning plans to restart their reprocessing plant, had been considering sand as a matrix material for treatment of cladding wastes before and after storage to prevent fines ignition. Sand has the advantage of being relatively free-flowing, and the maintenance problems are probably less than for other matrices. Metal recovery from a sand matrix, if desired, is also relatively simple.

B-3.2.2.2.2 Densification

B-3.2.2.2.2.1 Mechanical Compaction

The void fraction of untreated hulls, end adapters, grids, etc., is 80-90%.(2) ERDA 76-43 further estimates that compaction and flattening can at least triple the weight of hulls that can be stored in a given volume. As mentioned earlier in this section, there is an economic incentive for densification to optimize the handling, transport, and storage of the fuel bundle residues.

Unirradiated Zircaloy tubing has been compacted, and briquettes (27.9 cm diameter x 12.7 cm high), weighing 3.6 kg have been produced at about 72% of the theoretical density of Zircaloy.(2) Irradiated hulls may be embrittled and may fall apart when compacted, however.(2) Any fines so produced might increase pyrophoricity.

A commercially-available compacting unit could handle the hulls from a 5 MT/day spent fuel reprocessing plant and achieve 70% of the theoretical density.(2)

B-3.2.2.2.2.2 Melting

Densification by melting can reduce the fuel hull waste volume by a factor of about 6.(2) This represents a final volume about 40% less than that achieved by compacting to 70% of theoretical density. Zircaloy, stainless steel, and Inconel are currently produced using well-established melting technologies, though not in a radioactive environment. The melting of irradiated fuel hulls has yet to be demonstrated in quantity. The cost of melting would probably be higher than that of mechanical compaction.

B-3.2.2.2.2.3 Cold-Crucible Melting

The chemical activity of zirconium at its melting point of approximately 1850°C requires that it be melted in vacuum or an inert atmosphere. Conventional crucible materials react vigorously with molten zirconium. Consequently, cold-crucible processes were developed for producing high-purity zirconium.

The Inductoslag process, which was developed by the U.S. Bureau of Mines in Albany, Oregon, seems the most promising cold-crucible melting method for densification of fuel bundle residue. This process consists of induction-melting in a segmented, water-cooled, copper crucible. The system demands a high-purity slag, such as calcium fluoride. in an inert-gas atmosphere. The process has been demonstrated for the melting of simulated hulls. It produces a clean melt with a minimum of splatter. Disadvantages of the process are a relatively low production rate and the problems associated with slag handling and recycle.

Most densification studies with the Inductoslag process were concerned with the melting of decontaminated (descaled) fuel hulls potentially suitable for non-TRU waste storage or for reuse in the case of a segregated Zircaloy product. Melting of uncleaned, undescaled fuel hardware waste for the purpose of volume reduction alone may be desirable. Highly oxidized Zircaloy-4 was melted by the Inductoslag process in order to determine the ability of the process to densify uncleaned hulls. Melting characteristics were nearly identical with those of clean hulls.

B-3.2.2.2.2.4 Hot-Crucible Melting

Hot-crucible methods include ceramic crucible melting (for stainless steel or Inconel) and graphite crucible melting (for Zircaloy cladding or fittings). The major disadvantage of hot-crucible melting is the limited life of the crucible, necessitating remote replacement of crucibles and disposal of spent crucibles.

B-3.2.2.2.3 Conversion from Metal Form

B-3.2.2.2.3.1 Dissolution

Concentrated sulfuric and nitric acid mixtures have been used to dissolve hulls on a laboratory scale. Zircaloy dissolves rapidly, but dissolution of Inconel or stainless steel is slow. The dissolved TRUs could be separated and added to the main process stream in a reprocessing operation.

Similarly, fluoride solutions also attack Zircaloy rapidly, but are slow in dissolving stainless steel or Inconel. Little work has been done on the treatment of dissolved Zircaloy or on the separation of the dissolved TRUs.

B-3.2.2.2.3.2 Volatilization

In this process the chopped and leached fuel hulls are treated with a chlorinating agent (gaseous HCl or molten $ZnCl_2$) that disintegrates the hulls and hardware and converts them to metal chlorides. Since the zirconium chloride produced by this reaction is volatile, it can be separated from most of the other constituents of the primary wastes. The zirconium chloride could be reacted with steam to form zirconium oxide, which could then be incorporated in glass.

B-3.2.2.2.3.3 Oxidation with Hydrogen Fluoride-Oxygen Mixture

Fines of zirconium and stainless steel can be oxidized by hot air or oxygen. Larger pieces of Zircaloy or stainless steel can be burned in oxygen mixed with hydrogen fluoride gas. (The behavior of Inconel in hydrogen fluoride-oxygen mixtures at elevated temperatures is not known.) The resultant oxides would be suitable for incorporation into a glass.

B-3.2.2.2.4 Further Conversion of Nonmetallic Waste Forms

B-3.2.2.2.4.1 Conversion to Glass

Zirconium and iron-chrome-nickel alloy oxides are inert and thermodynamically stable and may be suitable forms for long-term storage. They do, however, have a large surface area that might be attacked by possible leaching solutions. Conversion of the oxides to a monolithic glass more resistant to leaching may therefore be desirable, although it increases waste volume.

B-3.2.2.2.4.2 Conversion to Adsorbents for High-Level Wastes (HLW)

Processes for making HLW adsorbents from zirconium have not been extensively investigated, although they are currently being studied at Sandia Laboratories. The ability to minimize leaching by adsorption and fixation of HLW ions on hydrous zirconium oxide is presently poorly defined and requires substantial development before it can be evaluated. The promise of volume reduction for the total waste is a primary motivating force for its evaluation. The volume of HLW calcine will be increased by adding the hull waste, however.

B-3.2.2.3 Evaluation of Alternatives

The alternatives described for the treatment of fuel hulls differ considerably in their state of development, but in each case the general character of the treatment product can be established. The operability of the processes is not so well established, nor are comparative costs. The available information on process description, status of technology and product description is summarized in Figure B-14 taken from ERDA 76-43.(2) Since demonstration experience and cost data are lacking, selections at this time tend to over-emphasize the product form. Nevertheless, the following can be noted from the Figure B-14 summary:

- Melt densification provides maximum volumetric efficiency, even with an allowance for some secondary waste resulting from slag recovery or decontamination.
- Zirconium in ingot form is highly resistant to corrosion, fire and leaching.
- Metal reuse may or may not prove to be economically feasible, but ingots melted from descaled hulls can be readily refabricated.

In summary, it appears that sorting to remove non-contaminated wastes, and decontamination are probably attractive cladding treatments. Sorting appears to have a strong economic incentive: this is not the case for decontamination under the proposed 10 nCi/g rule, but the principle of reducing the levels of TRU in the hulls and concentrating the residue for ease of handling is technologically sound. Revising the proposed 10 nCi/g rule might provide an economic reflection of this technical principle.

Volume reduction by compaction will probably be an economic advantage, but it does not reduce potential risks from waste disposal. Further stabilization by incorporation into concrete or glass matrices may be worthwhile, depending on the degree of integrity that could be achieved at reasonable cost. These stabilization technologies are not sufficiently developed at present, however, to permit optimization of waste form for these cladding wastes.

Although there are technological options for decontamination and/or increasing the leach resistance of cladding hulls, use of these options may not be cost-effective, in light of the relatively low activity of these wastes.

	PROCESS			STATUS OF TECHNOLOGY		PRODUCT DESCRIPTION							
TREATMENT ALTERNATIVE	REMOTELY OPERABLE	BATCH/SEMI-CONTINUOUS	MAINTENANCE EXPECTED	DETAILED PROCESS AVAILABLE	DEMONSTRATION LEVEL	LEACHABLE OR REACTIVE	VOLUME PRODUCTS/VOLUME HULLS	RESISTANT TO IGNITION	TRANSPORTABLE SAFELY	FURTHER TREATMENT POSSIBLE	SUITABLE FOR REUSE	STORABLE WITHOUT CONTAINMENT	COMMENTS
CONTAINED ONLY, NO TREATMENT	YES	B	LOW	YES	PLANT	YES	1/1	?	?	YES	NO	NO	CONSTITUTES PRESENT PRACTICE
MATRIX	YES	B	LOW	YES	--	YES	1+/1	YES	YES	YES	NO	NO	WHILE NOT DEMONSTRATED, ADDITION OF CONCRETE OR LOOSE SAND APPEARS TO BE WELL WITHIN CURRENT TECHNOLOGY
MECHANICAL COMPACTION	YES	C	?	NO	--	YES	1/3	?	?	NO	NO	NO	CONCEPTUAL ONLY - MECHANICAL PROPERTIES OF IRRADIATED FUEL HULLS LARGELY UNKNOWN
MECHANICAL COMPACTION AFTER DECONTAMINATION	YES	B, C	?	NO	--	NO	1/3+	?	YES	YES	NO	NO	AS ABOVE
DENSIFICATION BY MELTING	YES	B	?	YES	LAB	NO	1/6	YES	YES	NO	NO	YES	MELTING DEVELOPED THROUGH PROOF OF PRINCIPLE STAGE
DENSIFICATION BY MELTING WITH DECONTAMINATION	YES	B	?	?	LAB	NO	1/6+	YES	YES	YES	?	YES	DECONTAMINATION DEVELOPED THROUGH PROOF OF PRINCIPLE STAGE
DENSIFICATION BY MELTING WITH DECONTAMINATION SEGREGATION	YES	C	?	NO	LAB	NO	1/6+	YES	YES	YES	YES	YES	SEGREGATION OF FUEL HULL RESIDUE METALS HAS NOT BEEN DEMONSTRATED
DISSOLUTION IN SULFURIC ACID	YES	B	?	NO	LAB	YES	1/1	YES	?	YES	NO	NO	CAN HYDROLYZE TO OXIDE AND INCLUDE IN GLASS
DISSOLUTION IN AQUEOUS FLUORIDES	YES	B	LOW	YES	PLANT	YES	1/1	YES	?	YES	NO	NO	CAN HYDROLYZE TO OXIDE AND INCLUDE IN GLASS
FUEL HULL VOLATILIZATION	YES	B		NO	LAB	YES	1/1	YES	?	YES	YES	NO	CONVERSION TO HLW ABSORBENT
BURNING FUEL HULLS TO OXIDES	YES	C	?	NO	LAB	?	1/1	YES	?	YES	NO	NO	CAN BE INCLUDED IN GLASS
CONVERSION OF SIDE WASTE STREAMS	YES	B	?	NO	LAB	?	1/1	YES	?	YES	NO	NO	PROOF OF PRINCIPLE, TECHNIQUES FOR HANDLING SIDE WASTE STREAMS NOT DEMONSTRATED

FIGURE B–14 TREATMENT OF FUEL HULLS – PROCESS-PRODUCT CHARACTERISTICS AND STATUS

Source: Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle. ERDA 76-43, U.S. Energy Research and Development Administration, May 1976.

B-3.2.3 Storage Canisters

B-3.2.3.1 Canister Sizes

Canister sizes vary over a wide range of both diameter and length, depending on the heat-generation rate and the throughput of the process.

ERDA 76-43 gives the following probable criteria for HLW containers:

- Maximum diameter, 40 - 60 cm
- Maximum length, 3 - 4.5 m
- Maximum linear heat release rate, 2.9 kW/m
- Maximum radiation, 1×10^3 rem/hr neutron dose rate and 1×10^6 rem/hr gamma dose rate, measured 1 meter from any point on the surface of the canister
- Surface contamination not defined but must be low enough to meet allowable contamination levels at the Federal repository(2)

The above limits are based on factors that become restrictive after the solidified HLW leaves the reprocessing plant, i.e., during transportation and at the Federal repository. The limits on linear heat release rate and radiation, for example, can be exceeded when the canisters are initially filled.

In general, the diameter is controlled by the heat loading of the solidified product, the thermal conductivity, the heat removal capacity and the maximum acceptable surface and/or centerline temperature. Opinions vary on the limit that should be imposed on the maximum temperature. If devitrification of the glass would lead to a more leachable product, the temperature should be kept low enough to prevent crystallization. On the other hand, if certain additives are used to create insoluble crystalline phases, the above temperature limitation no longer applies.

The longer the fission products are cooled, either by storage of spent fuel elements before processing, or by storage of liquid wastes after reprocessing but before solidification, the larger the diameter of the canister that can be used. A ten-year cooling of fission products is assumed as the reference case (see Section B-3.3). Cooling for ten years will reduce the decay heat rate by a factor of about ten relative to the one-year value. Therefore, the discussion of heat dissipation from waste canisters is somewhat less important than would be the case with relatively fresh wastes. Nonetheless, heat dissipation efficiency does allow optimization of canister volume regardless of the waste specific heat.

Four possible schemes have been proposed for increasing the allowable heat generation in a canister. They are:

a) Use of annular inner containers,
b) Use of cylindrical canisters with internal cooling tubes,
c) Use of cylindrical canisters with internal fins, and
d) Incorporation of metals with the waste to give improved thermal conductivity.

These will be discussed below.

B-3.2.3.1.1 Annular Containers

Annular containers have been proposed for the British Rising Level Glass Process (HARVEST) because a high surface area for heat removal is needed as well as a large batch size. The design presently favored has a 1.22 m (48 in) o.d. and a 76.2 cm (30 in) i.d. with a 2.5 cm (1 in) wall thickness on both inside and outside shells. The canisters would be 2.74 m (9 ft) high and filled to 1.83 - 2.13 m (6-7 ft) with glass, and would contain approximately 1 m^3 of glass. The containers are designed for a glass with a maximum heat release of 140 W/liter and 140 kW total heat output per container.

An annular container for product storage is also proposed for the rotary kiln-ceramic melter unit for La Hague if the heat release is above 100 W/liter.

B-3.2.3.1.2 Containers with Internal Cooling Tubes

Internal cooling tubes have been proposed for cases in which the canister is filled by pouring glass directly from a melter. Each container would be fitted with seven tubes ranging in diameter from one-twelfth to one-fourth the diameter of the container. In this way, it is possible to increase the heat removal from a given outer container by a factor of 10 with some sacrifice in the filled volume.

B-3.2.3.1.3 Canisters with Internal Fins

Another method of increasing the heat removal from a cylindrical container is by fitting a number of fins inside the canister. In this way the heat removal rate can be doubled. The latest design leaves a 13 mm (0.5 in) gap between the fin and the wall of the canister. In this way, the wall temperature of the canister is more uniform and hot spots are avoided where the fins would otherwise touch the wall. The fin assembly is fabricated as a simple drop-in unit.

B-3.2.3.1.4 Canisters With High-Level Waste Solids Incorporated in a Metal Matrix

The incorporation of solidified wastes in metal matrices is discussed in Appendix B-II of this report.

B-3.2.3.2 Canister Materials

The selection of canister material is determined by the properties of the contained waste, by the solidification process, and by the desired degree of durability in the storage facility. We assume corrosion to be much more of a threat to canister integrity than stress caused by rock or salt movement in the short term. If the canister does not have to withstand a high temperature for more than a short time, as in the case of a ceramic-melter process in which the molten material is poured into the canister and cooling begins almost immediately, the properties of the glass may govern. For this situation where a relatively noncorrosive borosilicate-glass is produced, a carbon steel canister may be adequate. For cases where a high temperature must be sustained for a significant time, stainless steel may be required, even with a noncorrosive glass, because of its better structural ability to withstand the heat. This is the situation with an in-can melting process for example. Stainless steel may also be required for storing calcine where a relatively high-temperature bakeout period is employed. Because phosphate glass is corrosive to stainless steel, Inconel has been proposed for such service.

Resistance of the canister to corrosion in the geologic disposal medium may or may not be a critical factor, depending upon the degree of primary containment desired. To be conservative, it should be assumed that the canister will be in a hot, moist, salt-saturated environment. Pertinent corrosion data are presented in Appendix B-V.

Carbon steel and stainless steel have very poor corrosion resistance in hot brine. Although Inconel has outstanding resistance to corrosion by hot seawater at high velocity, it is quite vulnerable to pitting and serious localized attack in quiescent seawater.(15)

Titanium may be the best choice for highly leachable waste forms. Titanium and its alloys are outstanding in their resistance to seawater under all conditions of temperature and velocity.(15) Titanium corrosion rates in hot salt water are on the order of 0.002-0.02 mm/yr (0.1-1.0 mils/yr), the lower rate being for alloys, which corresponds to a lifetime of 1000-10,000 years for a 25 mm (1 in) canister.

It appears that carbon steel would be the best choice for canister material where high external corrosion rates may be tolerated. Titanium is the best material for long-lived canisters. Stainless steel might be the material of choice for glass forms because of the higher process temperatures, provided a rapid corrosion rate in the geologic medium is acceptable.

B-3.2.3.3 Post-Fill Canister Handling

The major post-fill operations required are seal-welding the canister and decontamination. Other operations shown in Figure B-15 can be employed for routine and non-routine quality control. These operations include calorimetry, radiation profilometry, and nondestructive measurement of wall thickness, and leak checking. Annealing of vitreous waste forms may also be included as a post-fill operation.

Although it is not presently envisioned as a requirement, it may be worthwhile to "overpack" the primary HLW canister with a secondary canister. Possible reasons for such overpacking include:

- Simplified decontamination. The secondary canister will not have been exposed to the contaminated environment of the processing cell.
- Improved quality of external containment. The secondary canister will not have been exposed to the high temperature (500 - 1050°C) of processing and will also have a lower differential expansion stress.

The disadvantages of overpacking include the extra costs involved and the requirement for reducing the heat release rate in the primary canister.

B-3.2.4 Off-Gas Treatment

Appendix B-III discusses off-gas control technology in detail. The following information is based on that Appendix.

Gaseous effluents from nuclear fuel reprocessing plants include chopper and dissolver off-gas and gases generated at the waste solidification facility. The chopper and dissolver operate at about 100°C. At this temperature, krypton, iodine, carbon dioxide, carbon monoxide, methane, nitrogen oxides, and some hydrogen and water vapor will all be evolved. At the higher temperatures of the solidification facility, there may be release of ruthenium and the remaining iodine. Nitrogen oxides are mainly a problem as interference with off-gas chemical processes, but all of the other items carry radioisotopes of concern: Kr-85, I-129, C-14, H-3 (tritium) and Ru-106.

Regulations (40 CFR 190) effectively require removal of about 80% of krypton and 99.6% of the iodine from the reprocessing plant off-gas. Regulations for C-14 control are being considered, but the level of removal that will be required is not yet known. No regulations exist for controlling tritium (H-3) or ruthenium, other than the site-boundary concentration limits of 10 CFR 20.

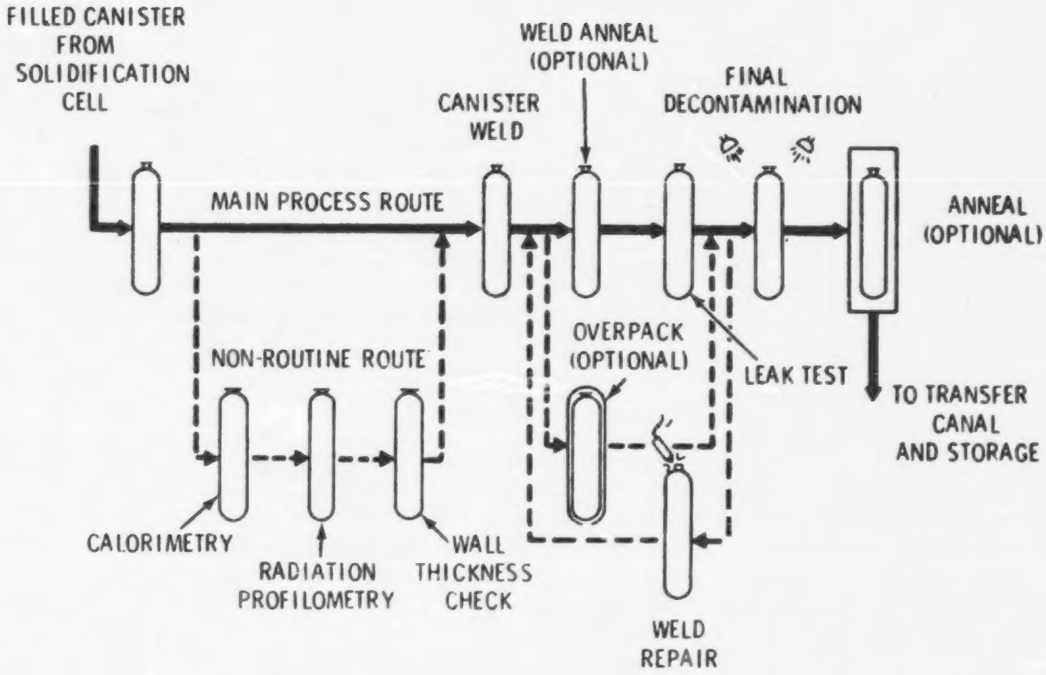

FILLED CANISTER FROM SOLIDIFICATION CELL
WELD ANNEAL (OPTIONAL)
FINAL DECONTAMINATION
CANISTER WELD
MAIN PROCESS ROUTE
ANNEAL (OPTIONAL)
NON-ROUTINE ROUTE
OVERPACK (OPTIONAL)
LEAK TEST
TO TRANSFER CANAL AND STORAGE
CALORIMETRY
RADIATION PROFILOMETRY
WALL THICKNESS CHECK
WELD REPAIR

FIGURE B–15 TYPICAL POST-FILL TREATMENT OF SOLIDIFIED HLLW CANISTERS

Source: Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle. ERDA 76-43, U.S. Energy Research and Development Administration, May 1976.

Krypton and iodine removal to meet the levels specified by 40 CFR 190 appears achievable with existing technology. Removal of carbon (mainly as CO_2) and ruthenium does not appear to be difficult, but the technology is not at the same level of development as for krypton and iodine removal. Tritium control is very difficult because it appears as HT gas and as HTO vapor and liquid, tending to seek isotopic equilibrium with H_2O streams.

Because removal of krypton and iodine from reprocessing plant off-gas streams is required, storage for them will have to be provided. The need for carbon and tritium removal and storage is still under discussion. Ruthenium removal and temporary storage may be required to meet site-boundary concentration limits.

For convenience of discussion, it will be assumed that the off-gas streams from the dissolver and from the HLLW solidification are combined. Therefore, a single removal facility (off-gas treatment plant) will be considered.

B-3.2.4.1 Krypton-85

Appendix B-III.1 discusses krypton control technology. It is clear from that discussion that krypton removal technology is available and will probably be incorporated into future reprocessing plants to meet the requirements of 40 CFR 190. Because Kr-85 has a short half-life (10.7 years), the most probable storage method is in pressurized bottles at the reprocessing site. Steel bottles in sheltered storage are capable of lasting at least 10 half-lives (enough time for the radioactivity to decay by a factor of more than 1000).

B-3.2.4.2 Iodine-129

Appendix B-III.2 discusses iodine control technology and concludes that the technology is available for removal of most of the iodine from reprocessing plant off-gas streams. If all the iodine in the fuel is released in the dissolver, then 99.6% removal is required to meet the requirement of 40 CFR 190, and technology is available to achieve this. Some iodine may, however, carry over into process streams and deposit throughout plant processing equipment, with an undetermined effect on the ability to meet the intent of 40 CFR 190.

The immobilization of I-129 will be necessary because of its extremely long half-life (1.6×10^7 years). Future storage standards may require a solid matrix with a large loading capacity, low leachability, nonflammability and simple handling characteristics.

Investigators at Oak Ridge National Laboratory have studied the use of cement for immobilizing iodine, and have found that 9% by weight iodine in the form of barium iodate can be incorporated into the matrix. This technology is designed especially to immobilize iodine collected by the Iodox process.

Use of silver and lead forms of zeolites for recovery and storage of I-129 has the advantage that collection and fixation of iodine occur in the same process. Additional immobilization of the loaded zeolite materials could be accomplished by the use of cements, glasses, or other bulk solidification methods.

B-3.2.4.3 Carbon-14

The need for removal of carbon-14 from the off-gas of a reprocessing plant (Appendix B-III.3) is still under study. Since most of the C-14 appears as CO_2, however, the removal technology itself would not be particularly difficult (CO_2 removal from air is a common industrial process).

The most obvious long-term disposal option for C-14 is to convert the wastes to calcium carbonate ($CaCO_3$) and package it in a container for burial in a deep geologic disposal site.

B-3.2.4.4 Tritium

Appendix B-III.4 discusses various tritium control technologies in detail, concluding that tritium removal from reprocessing streams is not technically achievable at present, although several methods may prove to be practical over the long term. It is not clear that tritium presents sufficient danger to warrant control. If, however, the tritium should be collected, then it may either be stored in tanks as tritiated water until sufficient decay has occurred to allow release, or it may be placed in cement for intermediate-term solid disposal.

B-3.2.4.5 Ruthenium-106

Appendix B-III.5 briefly discusses ruthenium control, pointing out that the release point will be at the waste solidification facility; the release mechanisms are poorly understood. Because ruthenium volatility is strongly dependent on temperature, it may be that ruthenium release at the solidification facility is more a process engineering problem (plateout and plugging) than a release problem. If it is determined that ruthenium constitutes a release problem, several technologies are possible for collection of the ruthenium, for example, adsorption on silica gel.

With a half-life of only one year, and substantial doubt as to whether much ruthenium-106 would be released from solidification processes, the ruthenium control problem seems not to be serious. Additional work needs to be done in this area, however.

B-3.2.5 Spent Fuel Assembly Disposal

Historically, it has been assumed that spent fuel assemblies, after some short-term storage, would be reprocessed. The valuable products, unburned uranium and the bred plutonium, would then be recycled into a subsequent batch of fuel, and the waste from this reprocessing operation would be solidified as described in previous sections of this report. If, however, a national policy decision is made to forego indefinitely the reprocessing of spent nuclear fuel, it will be necessary to dispose of the spent fuel assemblies without any prior reprocessing; this is the "throwaway fuel cycle."

To date, no criteria have been established for spent fuel disposal. The present practice of interim storage in pools at the reactor site or reprocessing plants will undoubtedly continue for some time. Possibly some "stand alone" spent fuel storage pools will be built to serve for the extended interim storage period. In this throwaway fuel scenario, however, a permanent disposal plan for spent fuel assemblies must be developed eventually. Although some estimates have been made of the costs of such permanent disposal, the literature seems essentially devoid of any account of engineering development of the required technology. Following are some general thoughts pertaining to preliminary concepts for disposal of spent fuel assemblies.

Three basic steps, with several sub-branches, must be considered in a total fuel assembly disposal cycle. Briefly they are:

1. Short- to Intermediate-Term Storage On-Site: High-density rack installations in existing pools or in a "new" independent pool to be constructed.

2. Intermediate-Term Storage Off-Site: Commitment to a storage facility that might be constructed by a commercial organization, by utilities (either individually or in a consortium) or by the Government.

3. Fuel Assembly Disposal: Shipment to a Federal facility for suitable preparation and geologic disposal, perhaps after a period of near-surface (retrievable) storage.

The cost and timing of each of these steps will be determined not only by the particular circumstances of the utility and its own plans for storage, but also by regulations yet to be established. Technical criteria are only now being considered for subsequent decisions that will lead to facilities to be constructed by industry and by the Federal Government. Each of these steps is discussed below, together with some of the considerations that bear on the timing and costs of the particular step.

B-3.2.5.1 Short-Term (Local) Storage

This first step in the disposal chain, as planned by utilities, consisted of a storage pool for spent fuel from the reactor. As originally conceived, storage would last only about six months, at which time fuel would be transferred from the reactor pool to a reprocessing plant. This plan of operation (including a "one-core spare" concept) set the pool size at about 1.3-1.7 reactor cores for a given plant.

When it became apparent that reprocessing would not be carried out as originally planned, most utilities elected to increase local storage capacity by substituting high-density fuel storage racks for the conservatively designed low-density concept. This approach represents a relatively short-term postponement of the problem, providing relief for up to ten years, depending upon the original pool configuration and type of reactor.

B-3.2.5.2 Interim Storage

Interim storage represents an undefined time period, beginning when the expanded short-term storage capability approaches its limit and continuing to the point where fuel assembly disposal (or at least long-term storage) under government control can be implemented. The final disposal or long-term storage has not yet been identified by the NRC, and until such definition the timing and actual handling procedures cannot be established. For many utilities it is clear that some interim storage requirement will be needed before the last disposal step. Several plans have been suggested for these alternatives.

From a technical standpoint, interim storage may be implemented either as an independent facility, or, if the timing of future nuclear installations at a particular site is suitable, as an expansion of the pool capacity of an additional unit planned or under construction. In this way, provision can be made for longer-term storage of fuel from the new unit, and for fuel from other units on that utility's system as well. Technically, either solution appears to be feasible, although an extra transportation leg is necessary with the independent facility (unless the storage site is located at the ultimate disposal site).

The fuel storage approach is assumed to use the "conventional" pool-type design that has been used at reactors and at reprocessing plants. Atlantic Richfield Company has proposed a totally different approach, in which after a storage period of up to 10 years, fuel assemblies are canned in nitrogen- or helium-filled containers and this canned assembly is then put into individual, relatively shallow, lined holes in the ground, with a sealed concrete cap for shielding. An array of these caissons or wells is spread over a relatively large area on about 7.6 m (25 ft) centers. The advantage of such a storage plan is that it does not require much maintenance or surveillance. It utilizes a totally passive heat removal system, with heat flowing from the caisson through the ground to the surface of the earth, using the atmosphere as an ultimate heat sink. Conceptually, this kind of an approach is halfway between interim storage and ultimate disposal; for longer-term storage it may have substantial merit. According to Atlantic Richfield, if the storage array is sufficiently large, the approach may be even less costly than conventional independent pool systems for interim storage.

B-3.2.5.3 Fuel Assemblies Disposal

Extensive investigation of permanent disposal for fuel assemblies is only now starting. A number of studies are being planned or are beginning; criteria and specifications may be developed within the next year or so.

The decision might be made to go to permanent deep disposal after a period of near-surface (retrievable) burial. The simplest of the fuel-disposal options involves canning the fuel assembly with an inert gas, such as nitrogen or argon. (After ten years of above-surface cooling, it is unlikely that it would be necessary to pack the spent fuel in a more efficient heat-transfer medium such as metal or a salt.) The canned assembly would then be placed in a mined-hole array like that proposed for high-level waste disposal canisters, or in deeper vertical holes drilled from the surface. The disposal area could be either salt or rock.

It is clear that the technology of disposal in salt is much further advanced than that of disposal in crystalline rock. Table B-9 summarizes the several alternative paths or decisions that may be implemented.

The above discussion assumes that the entire fuel assembly would be inserted in a canister, a gas added, and the canister sealed by welding. In the case of PWR spent fuel, a single assembly would be inserted in each canister. Since BWR fuel contains less fissionable material per assembly, two or three assemblies might be placed in a single canister. The canister cross section might be round, rectangular, or square. Round is simplest and would probably be least expensive from the standpoint of the canister alone. The subject of canister material has been explored in Section B-3.2.3.

TABLE B-9

FUEL ASSEMBLY DISPOSAL SUMMARY

Process Step	Time Frame	Implementation Alternatives
Short Term	Up to about 10 years after discharge.	Storage in own reactor pool (pool capacity increased to maximum extent). Storage in other reactor pool which has spare space.
Intermediate Term Storage	About 5-20 yrs. First independent facilities possible about 1982 with earliest disposal not before 1990.	An independent storage facility built by utility, government or third party. Independent dry storage facility (ARHCO proposal of wells in dry ground).
Fuel Assembly Disposal	R&D 1978-1982. Pilot operation 1982-1986. Disposal 1990 earliest.	Process Steps Involve: 1. Shipping 2. Canister Alternatives a. Carbon or stainless steel b. Titanium c. Double canning 3. Packing Alternatives a. Gas (N_2, He, Ar) b. Inorganic (S, NaCl) c. Metals (Pb, Al, Sn, Zn) 4. Disposal Alternatives a. Preliminary retrievable near-surface storage b. Deep Mine vs. Hole c. Deep Salt vs. Rock

B-3.3 SELECTION OF REFERENCE TECHNOLOGIES

It was shown in Section B-3.2 that many alternatives are being actively pursued for the treatment of high-level radioactive wastes. Some programs are ready for commercial application (spray, rotary-kiln, or fluidized-bed calcination; in-can or continuous melters). Alternative processes or products are being developed (supercalcine, sintering, metal matrix, glass-ceramic, coated-pellet, thermite, ion exchange).

The evidence from a technology assessment of waste solidification processes is that a selection could be made today from a number of processes that, assuming competent design, construction, and operation, would function satisfactorily for solidification of commercial reprocessing wastes. Existing defense wastes could also be accommodated, but unless the sodium nitrate content is removed extremely large volumes of solid would result.

Given the wide range of possibilities, the varied state of technological development, and the differing degrees of effectiveness for the alternatives, it is apparent that some weeding out of the less promising alternatives is essential in order to focus the evaluation effort on the most suitable techniques.

The approach used for this selection process has been to choose a set of reference cases that span a reasonable range of possibilities in terms of (a) waste form, (b) processing (solidification) approach, (c) packaging (containment) method, and (d) isolation effectiveness. In selecting reference cases, an attempt has been made to span a range of possibilities wide enough to permit extension to other combinations not specifically included in the reference cases.

In selecting reference technologies, the following criteria were used:

1. The reference case waste characteristics should span the reasonable range of possibilities available in the near term (within ten years).
2. The reference case technologies should be available for full-scale deployment for treatment of commercial reprocessing wastes.
3. Reference cases are based on U.S. technology, since there are several processes available in the United States that will meet commercial solidification needs.

Three solid waste forms have been selected for reference cases--calcine, glass, and spent fuel, all in suitable canisters. Disposal of waste as liquid has not been considered as a reference case because present regulations forbid leaving wastes as liquid over the long term.

(This should not be interpreted as precluding the possibility of direct disposal of liquid or slurry waste forms at some time in the future.)

B-3.3.1 Calcination

Early practical demonstration of waste solidification at the Idaho Chemical Processing Plant (ICPP) used the fluidized-bed calciner, which operated reasonably well in this capacity and has the most extensive operating history of any process in the United States. As detailed in Section B-3.2.1, however, some difficulties have been experienced in adapting this process to commercial reprocessing wastes. Based on an extensive development program that has included commercial-sized facilities, Battelle Northwest Laboratory (BNWL) favors the simpler spray calciner for commercial application, an opinion that appears sound. Therefore, the reference calcination process chosen is spray calcination. Because the spray calciner product is a finer powder than the product from any other processes, conservative evaluations of the leaching behavior will result.

B-3.3.2 Glassification

The reference case for glassification is taken to be in-can melting rather than joule-heated melting, because of its more extensive demonstration history with radioactive wastes and the simplicity of the process operation, which does not require transfer of wastes from a separate melter vessel.

Borosilicate glass is assumed as the reference glass because it is universally the most accepted type.

B-3.3.3 Spent Fuel Disposal

Section B-3.2 defines a number of options for disposal of spent fuel directly as waste. Although disposal options for spent fuel could include an intermediate processing step to reduce the spent fuel to a modified form (e.g., glass), such options have not yet been seriously studied, as the concept of "throwaway" fuel is very recent. The simplest option for spent fuel disposal, and the most attractive in terms of possible intermediate retrievable storage,is packaging in canisters with an inert gas blanket. Although design optimization might result in use of metal packing (for efficient heat removal) or double-canning, the simplest case is taken as the reference--i.e., single-canning with a nitrogen blanket.

B-3.3.4 Canisters

Each of the above-defined reference waste treatment processes (spray calcination, in-can melting, direct spent fuel disposal using nitrogen blanket in a single can) requires a protective canister.

It was pointed out in Section B-3.2.3 that carbon steel is the least expensive choice for canister material, but that it also will not last long in a wet, warm, salt-saturated environment. Stainless steel is not expected to be any more corrosion-resistant than carbon steel and, in fact, might be worse. Inconel has exhibited pitting characteristics, which put its survivability in question. Titanium is the only practical metal with reasonable probability of long-term (up to 1000 years) life in wet, hot salt. It was also pointed out that the need for long canister life is less in the case of glass than in the case of calcine.

In the reference cases, calcine product is assumed to be placed either in carbon steel cans or in titanium cans. Because calcine is the most leachable of the solidified HLW products, the carbon steel can provide a worst-case bound on waste-disposal scenarios.

The reference canister for glass forms is taken to be stainless steel, on the basis that in-can melting temperatures may create problems with carbon steel cans. Use of a titanium can would be of little advantage when used with the glass form, which has a very low leach rate.

Two reference cases are assumed for spent fuel canisters--again, carbon steel and titanium. Because of the possibility that spent fuel might be highly leachable, this pair of cases provides a test of the sensitivity of high-leachability forms to canister corrosion rates.

B-3.3.5 Off-Gas Residue Disposal

Volatile wastes released to the reprocessing plant off-gas system (either from the chopper/dissolver or from the solidification step), may need to be collected and stored prior to disposal. Section B-3.2.4 and Appendix B-III show that the volatiles of concern are krypton-85, iodine-129, carbon-14, tritium, and perhaps ruthenium-106.

Krypton removal by either cryogenic distillation or fluorocarbon absorption is feasible, and there are no significant barriers to application at commercial reprocessing plants. In either case, the resultant product will be krypton gas (mixed with quantities of other gases, depending on the technique selected). Although several "exotic" krypton stabilization techniques are being investigated (zeolite or clathrate entrapment), it is expected that the storage method finally selected will be containment in carbon steel cylinders, which are expected to have a lifetime far in excess of that needed to ensure decay of the krypton to very low levels.

Iodine removal is feasible and available for commercial application; the Mercurex process followed by silver zeolite polishers was selected for application at Allied General Nuclear Services (AGNS). The silver zeolite polisher concept is conceptually simple and may achieve almost any degree of decontamination desired, by addition of more polishers. The Mercurex process competes with the Iodox process for preliminary iodine removal (see Appendix B-III.2 for details); since the Mercurex process generates the larger volume of waste, it will be conservatively assumed that the Mercurex process is used for the treatment ahead of the silver zeolite beds. The waste form assumed for the Mercurex process is mercuric iodate. The simplest long-term disposal procedure would then be simply packing the mercuric iodate and spent silver zeolite in steel canisters.

Carbon-14 will mostly be in the form of carbon dioxide, which may be removed by a number of processes that yield calcium carbonate as their common product. This could then be placed in steel canisters for ultimate disposal.

Tritium control does not appear to be feasible or necessary at present, and the reference case is, therefore, taken to be release. The half-life of ruthenium-106 is so short that no reference case is presented.

B-3.3.6 Cladding Hulls and Fuel Bundle Residues

The reference case for disposal of fuel cladding hulls and fuel bundle residues is based on proposed regulatory framework implicitly requiring geologic disposal of wastes containing greater than 10 nCi/g TRU contamination. Such a regulation provides strong economic incentive (discussed in Section B-3.4) for volume reduction to reduce geologic disposal costs. The proposed 10 nCi/g TRU rule, therefore, encourages sorting of fuel bundle residues in order to reduce the volume of contaminated wastes that must be dealt with. This process step is particularly attractive if, as appears to be the case, the contaminated and non-contaminated wastes are relatively simple to separate--in this case, by mechanical means.

On the other hand, the 10 nCi/g rule would probably discourage the addition of a decontamination step to the cladding treatment, since the reprocessor will achieve no cost savings for reducing the amount of TRU contamination present in the cladding, as long as it is contaminated to higher than 10 nCi/g. (This may be something of an oversimplification, since decontamination may allow reductions in handling costs at subsequent processing steps.) Although decontamination appears logical (since it may concentrate most of the contamination for greater ease of handling and immobilization), the reference case assumes no decontamination, reflecting what appears to be the most likely actual process.

Because volume reduction appears to carry with it economic rewards through reduced costs for geologic disposal, a simple mechanical volume reduction step is assumed (compaction). Compaction is estimated to achieve three-fold volume reductions. Because no practical melting technology is at hand, only mechanical compaction is assumed.

In Section B-3.4 it will be pointed out that there may be a need for further definition of TRU wastes into low- and high-level categories in order to delineate more precisely the actual level of risk, instead of treating identically all wastes contaminated to greater than 10 nCi/g. What may be called for is a risk assessment that considers lower contamination levels and higher stability of form. Were such a program implemented, then further treatments of cladding waste (matrix formation in glass, sand or concrete, or conversion to non-metals) could be considered as part of this optimization process. As an example of this type of trade-off, it is not clear whether, under a sliding-scale TRU regulation, decontamination of the hulls followed by solidification of the decontamination residue and packaging of the hulls for low-level storage would be better than solidification and disposal of the undecontaminated hulls directly.

The reference case used is disposal of chopped hulls, sorted to remove end fittings, grids, and guide tubes, compacted to about one-third of the original volume, and packed into carbon steel containers for ultimate disposal. This reference case, based upon proven technology, is conservative insofar as it produces a waste form that could be improved upon by more advanced treatments.

B-3.3.7 Summary of Reference Cases

Following are the assumed reference cases for HLW disposal:

- Calcine: spray calcination, calcine contained in carbon steel or titanium cans for deep disposal.
- Glass: in-can melt, glass contained in stainless steel cans for deep disposal.
- Spent fuel: nitrogen-blanketed in carbon steel or titanium cans for deep disposal.
- Cladding residue: mechanically sorted from end pieces, compacted to one-third the original volume, placed in carbon steel cans for deep disposal.

In addition, for associated wastes, reference cases are:

- Krypton gas storage at the reprocessing plant in carbon steel gas cylinders.

- Iodine recovery by Mercurex process followed by silver zeolite beds, the mercuric iodate and spent zeolite packed into carbon steel canisters for deep disposal.
- Carbon-14 recovery as calcium carbonate contained in carbon steel canisters for disposal.
- Tritium release during reprocessing, or from spent fuel as the fuel is leached.

B-3.4 DISPOSAL OF OTHER TRU-CONTAMINATED WASTE

Task A described the nature of low- to intermediate-level transuranic-contaminated (TRU) wastes that originate in the nuclear fuel cycle and estimated the possible quantities of TRU waste arising from various fuel cycle scenarios. The subject of TRU waste treatment covers a broad range because of the wide variety of waste characteristics. Wastes may include general process trash (rags, plastic, paper, tools, glass and metal), failed or expended equipment (machines, valves and glove boxes), and liquids (slurries, sludge, resins, oil, and grease). In this section, the technologies for treatment of these wastes will be discussed.

As was pointed out in Task A, considerable uncertainty exists concerning the physical characteristics of TRU waste arising from various stages of the fuel cycle. A degree of uncertainty still exists about the ultimate disposition of TRU waste, although the regulatory thrust seems to be in the direction of requiring geologic disposal of waste contaminated to levels above 10^{-8}Ci/g. If this proposed "10 nanocurie rule" is adopted, there will be considerable economic incentive to reduce TRU waste volume. The fact that ERDA's New Mexico salt project is primarily designed to accommodate low-level TRU waste, albeit retrievable,(16) is indicative of the trend toward geologic disposal of TRU wastes. The current Office of Waste Isolation (OWI) commercial radioactive waste disposal program also includes provisions for TRU waste disposal.(17)

As an illustration of the economic incentive for TRU waste volume reduction, cost figures to be given in Section B-4 for a large low-heat-generation geologic waste disposal facility will show that for every cubic meter of TRU waste volume reduction, there is a potential cost saving of at least $860. It is thus not surprising that a comprehensive program is under way to develop volume reduction techniques for handling TRU waste.(18)

TRU waste, although it takes many forms, may be classified into three categories for purposes of discussing treatment methods: combustible, non-combustible, and liquids/sludges, which would be evaporated to a sludge or cake and fixed in a solid such as concrete. Figure B-16 shows an overview of possible TRU waste material flows through the various waste treatment stages.(18)

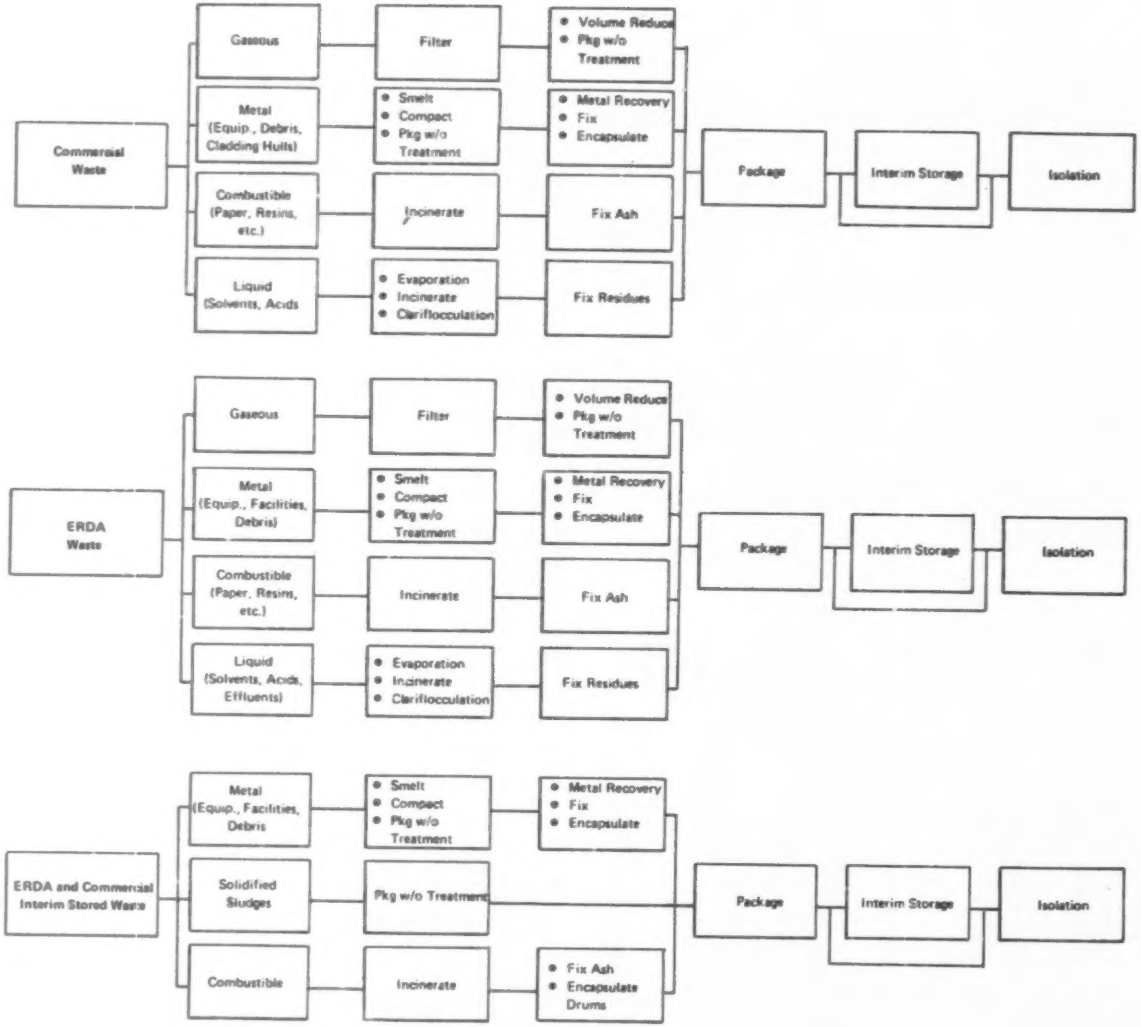

Commercial Waste
Gaseous
Metal (Equip., Debris, Cladding Hulls)
Combustible (Paper, Resins, etc.)
Liquid (Solvents, Acids
Filter
• Smelt • Compact • Pkg w/o Treatment
Incinerate
• Evaporation • Incinerate • Clariflocculation
• Volume Reduce • Pkg w/o Treatment
• Metal Recovery • Fix • Encapsulate
Fix Ash
Fix Residues
Package
Interim Storage
Isolation
ERDA Waste
Gaseous
Metal (Equip., Facilities, Debris)
Combustible (Paper, Resins, etc.)
Liquid (Solvents, Acids, Effluents)
Filter
• Smelt • Compact • Pkg w/o Treatment
Incinerate
• Evaporation • Incinerate • Clariflocculation
• Volume Reduce • Pkg w/o Treatment
• Metal Recovery • Fix • Encapsulate
Fix Ash
Fix Residues
Package
Interim Storage
Isolation
ERDA and Commercial Interim Stored Waste
Metal (Equip., Facilities, Debris
Solidified Sludges
Combustible
• Smelt • Compact • Pkg w/o Treatment
Pkg w/o Treatment
Incinerate
• Metal Recovery • Fix • Encapsulate
• Fix Ash • Encapsulate Drums
Package
Interim Storage
Isolation

FIGURE B–16 MANAGEMENT METHODS FOR TRANSURANIC-CONTAMINATED WASTES

Source: Wolfe, R.A. The Research and Development Program for Transuranic-Contaminated Waste Within the U.S. ERDA. In: Radioactive Wastes from the Nuclear Fuel-Cycle AIChE Symposium Series 154:72, 1976.

TRU waste is mostly "low-level TRU waste", which means that the gamma radiation is sufficiently low to allow handling of packaged wastes without shielding (less than 10 mrem/hr surface radiation). These wastes are characterized(19) as having an average plutonium content before compaction of 9 g/m^3, corresponding to a plutonium processing loss of about 1%. Also generated will be "intermediate-level TRU waste", which is characterized as having sufficient external radiation (10-1000 mrem/hr) to require some shielding and special handling, but a lower average plutonium content (1 g/m^3, corresponding to a 0.025% plutonium loss). Because this intermediate-level TRU waste appears in lower quantities than low-level TRU waste (see Task A), it may be possible to eliminate shielding requirements for intermediate-level TRU waste by combining the wastes, and thereby achieve self-shielding sufficient to reduce surface radiation levels.

A first step toward reduction of TRU waste volume is minimizing the waste generation from fuel cycle processing steps. Waste minimization may be accomplished both by the design of a facility and by administrative controls, such as procedures that minimize the production of analytical or clean-up waste. Facility design can contribute to reduction of waste quantities by increasing chemical concentrations in process streams, recycling waste streams, and optimizing ventilation flow rate controls to increase filter life in dirty conditions. Administrative controls include such techniques as recovering spills with solution chemistry rather than rags and towels, using corrosion-resistant tools, and decontaminating the waste itself (e.g., tool washdown and re-use). Assay and sorting may achieve further volume reduction by releasing wastes with less than 10 nCi/g. At upwards of \$860/m^3 disposal costs, a sorting/decontamination operation could perhaps be economically justified.

After steps have been taken to 1) reduce primary waste quantities; 2) decontaminate waste to return some of it to unrestricted use; and 3) sort the contaminated waste into combustibles, non-combustibles, and liquids, steps may be taken to reduce waste volume, and to stabilize and package the waste.

B-3.4.1 Combustibles

The simplest treatment for combustibles is compaction, which is also inexpensive. In Task A it was shown that a four- to five-fold volume reduction may be achieved. Compaction of combustibles is probably not the best solution, however, because it leaves two potential problems with the waste: fire hazard, which is a remote possibility, especially if the compacted wastes are packaged in metal drums or other fire-resistant packages; and radiolysis of waste hydrocarbons, releasing hydrogen.

Incineration appears to be the most promising method of treating TRU wastes. Six potentially attractive incineration methods--controlled-air, pyrolysis, molten salt, fluidized-bed, cyclone, and acid digestion--have been identified and discussed in the literature.(18,20,21,22) The state of development of these six techniques is shown in Figure B-17.(18) A brief description of each method follows.

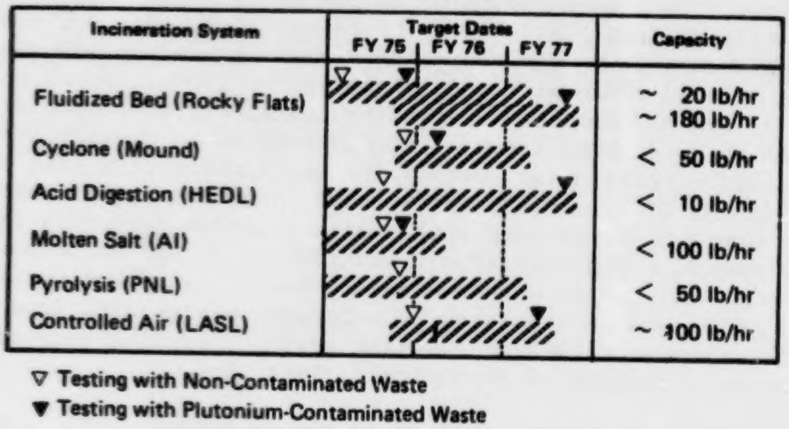


FIGURE B–17 DEVELOPMENT OF INCINERATION METHODS FOR TRANSURANIC-CONTAMINATED WASTES

Source: Wolfe, R.A. The Research and Development Program for Transuranic-Contaminated Waste within the U.S. ERDA. AIChE Symposium Series 154:72, 1976.

B-3.4.1.1 Controlled-Air Incineration(18,22)

Los Alamos Scientific Laboratory (LASL) is building a 45 kg/hr (100 lb/hr) developmental incinerator using an adaptation of standard municipal incinerators, with a combustion and off-gas burning chamber to achieve complete combustion. Off-gases will be scrubbed and liquid waste will be sent to a disposal facility. Although a volume reduction factor of 20 is conservatively predicted,(23) LASL has informally estimated a net volume reduction factor of 30 from primary waste after inclusion of secondary wastes arising from the incineration system.

B-3.4.1.2 Pyrolysis(18,22)

The pyrolysis-burning concept (heating in an oxygen-deficient atmosphere, to gasify part of the waste material, which is then combined with air and burned in a secondary combustion chamber) is being developed at BNWL at Richland, Washington, and is an adaptation of systems used to treat industrial waste. The process achieves a significant volume reduction by incineration. The process has been demonstrated with non-radioactive simulated waste at 15 kg/hr (33 lb/hr).

B-3.4.1.3 Molten Salt Combustion(18,20,22)

The molten salt combustion process is being developed by Atomics International Division of North American Rockwell Corporation at Canoga Park, California. Waste volume is reduced by utilizing molten sodium carbonate or lithium carbonate as a medium to burn organic materials, trap particulates, and react chemically with any acidic gases produced during combustion. The radioactive materials are contained in the molten salt. The melt-ash mixture is processed to separate ash for disposal, to recover the salt for recycle, and potentially to recover plutonium. Combustion has been demonstrated at 0.5 kg/hr (1 lb/hr) on a bench scale with wastes contaminated with plutonium and beta-gamma activity, and on a pilot scale with uncontaminated waste.

B-3.4.1.4 Fluidized-Bed Combustion(18,22)

A fluidized-bed combustion system is currently being installed in a plutonium recovery facility at the Rocky Flats nuclear defense plant, operated for ERDA by the Rockwell International Company. A prototype unit was tested with noncontaminated waste for about a year and became operational about May 1975 for the recovery of plutonium-contaminated scrap materials. This prototype system utilizes a fluidized combustion bed, a fluidized catalytic afterburner, and a porous metal filter. The system has a continuous capacity of about 10 kg/hr (22 lb/hr). A demonstration plant rated at 82 kg/hr (180 lb/hr) is being built.

B-3.4.1.5 Cyclone Incinerator(18,22)

The cyclone incinerator concept has been tested at the ERDA Mound Laboratory operated by the Monsanto Research Corporation in Miamisburg, Ohio. This system utilizes the concept of igniting waste within a metal drum container while injecting air in a vortex pattern to effect complete combustion. The gaseous effluents are passed through a self-cleaning fiber-bed filter system. The water used for scrubbing the filter will be neutralized and recycled within a closed-loop system. The demonstration unit being tested has a capacity to incinerate up to 23 kg/hr (50 lb/hr) of plutonium-contaminated combustible waste.

B-3.4.1.6 Acid Digestion(18,21,22)

This process is being developed at Hanford Engineering Development Laboratory (HEDL) to reduce the volume of combustible waste by digesting it in hot (230-270°C) concentrated sulfuric acid containing nitric acid oxidant to form noncombustible residues having less than 40% of their original volume. The residue is also in a form that could provide for recovery of the plutonium. HEDL has demonstrated this process at 0.5 kg/hr (1.1 lb/hr)with nonradioactive waste. A radioactive waste demonstration unit of the same size is being constructed.

B-3.4.1.7 Summaries of Incineration Development

Cooley and Clark catalogue incineration processes, including vortex, agitated-hearth, and moving-belt approaches.(22) Table B-10 lists incinerators that have been or are being used for combustion of radioactive solid wastes; Table B-11 lists processes under development. Perkins has provided a further review.(23) The most promising techniques appear to be the LASL controlled-air and the Rocky Flats fluidized-bed processes.

B-3.4.2 Non-Combustibles(18,22)

After they are decontaminated and sorted to reduce waste quantity to a minimum, non-combustibles may be compacted or smelted to reduce volume. Compaction may consist of mechanical disassembly and/or compression with a heavy compactor. Depending on radiation levels, this may be accomplished by contact maintenance (Rocky Flats uses a special room with maintenance personnel in pressurized suits) or by remote maintenance for higher-gamma radiation.

Smelting is under development and has the advantages of immobilization of waste (decreasing the surface-to-volume ratio) and volume reduction.

TABLE B-10

INCINERATORS USED IN THE UNITED STATES FOR THE COMBUSTION OF RADIOACTIVE SOLID WASTES

Incinerator installation	Location	Year built	Operational status	Operating features	Capacity (kg/hr)
Knolls Atomic Power Laboratory	New York	1949	Not in operation	Single chamber; pure O_2; 550°C	9 to 18
Argonne National Laboratory	Illinois	1951	Not in operation	Single chamber; 450 to 600°C; vibrating grate; gas fired	68
Bettis Atomic Power Laboratory	Pennsylvania	1953	Not in operation	Single chamber; cyclone air feed	–
Shippingport Atomic Power Station	Pennsylvania	–	Not in operation	Cyclone air feed	18
United Nuclear Corporation	Connecticut	~1960	Not in operation	–	95
Metals and Controls	Maine	~1960	Not in operation	–	90
Pratt and Whitney Aircraft Company	Connecticut	~1960	Not in operation	–	270
Douglas United Nuclear	Washington	1967	Not in operation	Natural draft incinerator for open-pit burning	–
Yankee Rowe Atomic Electric Company	Maine	~1968	In operation	Cyclone air feed	18
National Lead Company of Ohio	Ohio	1954	In operation	Dual chamber; 980°C; gas fired	1000
Union Carbide K-25 Plant	Tennessee	1972	In operation	Dual chamber; 930 to 1100°C; gas fired	–
Union Carbide–Paducah	Kentucky	–	Not in operation	–	–
General Electric Company, Atomic Power Equipment Department	California	1960	Not in operation	Triple chamber; 760 to 820°C; gas fired	45
Goodyear Atomic Corporation	Ohio	1971	In operation	Dual chamber; 815 to 1000°C; gas fired	68
Gulf General Atomics	California	1963	In operation	Dual chamber; 900 to 1200°C; gas fired	20
Nuclear Fuel Services, Inc.	Tennessee	–	In operation	Dual chamber; gas fired	270
Union Carbide Y-12 Plant	Tennessee	1955	In operation	Single chamber; 870°C; gas fired	~20
Babcock & Wilcox	Virginia	1972	In operation	Single chamber; 1090°C	80
Los Alamos Scientific Laboratory, plutonium-recovery incinerator	New Mexico	1952	In operation	Single basket; 800°C; electrically heated	<1
Mound Laboratory, plutonium-recovery incinerator	Ohio	~1972	In operation	Single basket; 800°C; electrically heated	23
U.S. Army Nuclear Defense Laboratory	Maryland	1963	Not in operation	Dual chamber; cyclone air feed	23

TABLE B-10
(continued)

Incinerator installation	Location	Year built	Operational status	Operating features	Capacity (kg/hr)
Kerr-McGee Nuclear Corporation	Ohio	1972	In operation	Dual chamber; gas fired	70
Los Alamos Scientific Laboratory (development)	New Mexico	(1976)	Under construction	Dual chamber; 500 to 800°C; controlled air	45
Mound Laboratory (development)	Ohio	1975	In operation	Single chamber; cyclone air feed; 1100°C	35
Rocky Flats Plant (development–agitated hearth)	Colorado	(1980)	Under construction	Agitated hearth; 800 to 1000°C; fired with diesel fuel	70
General Electric Nuclear Fuel Plant	North Carolina	1972	In operation	Vortex burner; 815 to 980°C; gas fired	450
Westinghouse Nuclear Fuel Division	South Carolina	1974	In operation	Dual Chamber; 650 to 1200°C; excess air burner	–
United Nuclear Corporation	Rhode Island	1967	Not in operation	Single chamber; gas fired	45
Rocky Flats Plant (development–rotary kiln)	Colorado	(~1980)	Under construction	Rotary kiln; 600 to 800°C; fired with diesel fuel	40
Rocky Flats Plant	Colorado	1959	In operation	Dual chamber; 1200 to 1400°C	16
Babcock & Wilcox Nuclear Materials Division	Pennsylvania	1969	Not in operation	Batch burner; dual chamber; gas fired	9
Atlantic Richfield Hanford Company plutonium-recovery incinerator	Washington	1961	In operation	Dual chamber; 700 to 800°C; moving grate	2

Source: Cooley, C.R. and D.E. Clark. Treatment Technologies for Non-High-Level Wastes (USA). In: Proceedings of the International Symposium on the Management of Wastes from the LWR Fuel Cycle. CONF 76-0701, U.S. Energy Research and Development Administration, 1976

TABLE B-11

PROCESSES UNDER DEVELOPMENT IN THE UNITED STATES FOR THE COMBUSTION OF RADIOACTIVE SOLID WASTES

		Development status				
				Radioactive unit		
Process	Noncombustible product	Site of development	Tested capacity (kg/hr)	Design capacity (kg/hr)	Scheduled startup	Commercial availability
Controlled-air incineration	Highly refractory oxide	Los Alamos Scientific Laboratory	45	45	1977	1978
Cyclone incineration (excess air)	Highly refractory oxide	Mound Laboratory	35	35	1978	1980
Fluidized-bed incineration	Refractory oxide	Rocky Flats	9	82	1977	1980
Rotary-kiln incineration	Refractory oxide	Rocky Flats	2	40	1980	1982
Agitated-hearth incineration	Refractory oxide	Rocky Flats	4	70	1980	1982
Controlled pyrolysis	Refractory oxide	Pacific Northwest Laboratories	15	None	[b]	[b]
Molten-salt combustion	Salt-ash or an oxide if salt is leached	Atomics International	50	50	1978	1978
Acid digestion	Sulfates and oxides	Hanford Engineering Development Laboratory	5	5	1977	1980

---

[a] All processes lead to volume-reduction factors in the range of 20 to 40, assuging the same mixed waste for each process and initial processing only (i.e., no final conversion of residue nor secondary waste generated by the process itself).

[b] Not currently funded; estimated 3 years needed to develop process for commercial application.

Source: Cooley, C.R. and D.E. Clark. Treatment Technologies for Non-High-Level Wastes (USA). In: Proceedings of the International Symposium on the Management of Wastes from the LWR Fuel Cycle. CONF 76-0701, U.S. Energy Research and Development Administration, 1976

B-3.4.3 Wet Wastes (Liquid and Sludges)(18,22)

There are a number of options for treatment of wet wastes: evaporation, drying or calcination for high-solids content; filtration or ion exchange for low-solids content; combustion for combustible liquids.

These techniques are widely used for both radioactive and nonradioactive industrial processes, and the choice of technique will depend on design compatibility and economics. The resultant residue will be a solid, either salt cake, resin or calcine. Reverse osmosis or flocculation may be used as a concentrating mechanism for low-solid liquids, but a second drying or filtering step must be used to solidify the resultant concentrates.

B-3.4.4 Final Packaging

TRU waste is now placed in crates or drums. The crates are fiberglass-coated plywood with polyvinylchloride (PVC) and fiberboard liners built to meet Department of Transportation Specification DOT-7a. The plastic-lined, 208-liter (55-gal) drums are built to meet Department of Transportation Specification DOT-17c.(25) These packages are suitable for retrievable storage. For ultimate TRU waste storage, these retrievable-storage drums or crates may be used, or the wastes may be transferred to a container that is easier to handle in a geologic disposal facility.

Although TRU waste is presently defined simply as waste containing greater than 10 nCi/g transuranic contamination, in fact there will be a wide range of contamination levels, up to perhaps several million nCi/g on some ventilation filters. The options ultimately selected for fixation of these TRU wastes should reflect this range of contamination, with higher contamination levels subjected to high levels of fixation. It may, for example, be desirable to fix higher-level wastes in concrete, whereas drums may be satisfactory for lower levels. In general, the work necessary to define these options has not yet been performed. There is considerable experience at ORNL, however, in using concrete and grouts to fix intermediate-level wastes.(26) In addition, Atlantic Richfield Hanford Company (ARHCO) and BNWL have investigated placing alpha wastes in glass on a laboratory scale, successfully converting certain types of waste to immobile silicate glasses.(27)

B-3.5 ALTERNATIVE GEOLOGIC DISPOSAL TECHNIQUES

Disposal of radioactive wastes in deep, stable geologic formations has long been considered the preferred method for isolation of these wastes from contact with man's environment. A number of possible geologic media have been considered for such disposal; these include salt beds, salt domes, crystalline rock forms such as granite or basalt, shales, limestones, certain types of clay beds, and others. To date salt deposits have received the most attention, especially in the

United States, because of their demonstrated stability over very long time periods, their homogeneity, and their capability of plastic flow (self-healing) in the presence of stress. The self-healing characteristics of salt effectively eliminate the possibility of extensive cracking, thereby preventing the opening of pathways to the environment.

An alternative to salt is a stable crystalline rock, such as basalt or granite. Again, there are abundant examples of suitable depth and age with demonstrated seismic stability. Crystalline rock does not have the self-healing characteristics of salt, but possesses other advantages, e.g., resistance to water intrusion, that make it a desirable medium for geologic disposal of radioactive waste.

In addition to salt and crystalline rock, other geologic media have been considered, e.g., shales and clay deposits. In general, these have both desirable and undesirable characteristics. For example, the laminar structure of horizontally-bedded shale reduces water permeability in the normal (vertical) direction, but the presence of water could lead to high stresses and possible disintegration of the shale at high temperatures. Certain types of clay-till deposits also have the advantage of low water permeability, but the disadvantage of indeterminate long-term stability characteristics.

In light of the foregoing, the following have been selected as reference cases for the purposes of this study:

(a) Disposal in bedded salt
(b) Disposal in granite
(c) Disposal in salt domes

A uniform depth of 460 m (1500 ft) has been assumed for all three cases.

The design of disposal facilities for radioactive wastes generally follows conventional practices with two important exceptions: (1) special provisions are required for safe transport and cooling of radioactive material; and (2) waste canister spacing and arrangement must be in accordance with thermal design criteria for the specific medium. These criteria generally take the form of bulk temperature limitations and/or temperature gradient limitations to ensure reasonable integrity of the repository. Because of their importance, these thermal criteria will be discussed in considerably more detail in Section B-3.5.3

Until recently, salt was the leading contender for geologic disposal, but more attention is now being focused on other media, such as crystalline rock. Despite growing interest in these other media, nearly all of the work performed to date on engineering design of disposal facilities has been for salt deposits; the information available for media other than salt is extremely limited. Therefore, the major focus of this section of the report will be on salt deposits, principally bedded salt.

B-3.5.1 Design Characteristics for Disposal in Bedded Salt

The reference design described below assumes non-retrievable underground mine storage in a bedded salt deposit. The state of the art for the technology required to develop such a facility is assessed and areas where further study is required in order to optimize waste handling and facility design are noted. Fully retrievable storage and storage in other media, such as granite or shale, are also discussed briefly.

Since it is not clear at present whether reprocessing of spent fuel will be resumed, two reference waste burial facility designs will be considered in this section. The first design is based on storage of the wastes from the reprocessing of plutonium-recycle fuel. The second design is based on storage of spent fuel and other wastes from a throwaway fuel cycle.

The two facility design concepts are similar. Each will have a surface facility; a series of shafts to transport personnel, equipment, and wastes to the mine level; and segregated storage locations in the mine for storage of HLW or spent fuel, cladding wastes, intermediate-level TRU wastes, and low-level TRU wastes. Major differences are in the equipment dimensions, mine corridor dimensions, waste canister spacing, and the overall number of waste units to be buried. Because of the basic similarity for the two reference facilities, the following discussion will concentrate on the design of a facility to bury wastes from a reprocessing fuel cycle. The major design differences for the throwaway fuel cycle burial facility will be noted at the end of this discussion.

The reference design for this study assumes burial in a mine located 460 m (1500 ft) below the surface. The facility's general arrangement is similar to the waste isolation facility developed under contract to the Office of Waste Isolation (OWI).(17) Various components of the facility will be discussed in more detail below.

The reference facility is sized to accept ten-year-old wastes from reprocessed equilibrium mixed-oxide fuel or ten-year-old spent fuel from a throwaway fuel cycle. The ten-year delay between fuel discharge and waste burial is arbitrary. NRC regulations require that all HLW from a reprocessing stream be solidified within five years and sent to a Federal repository within ten years. With older fuel, there are handling advantages, such as reduced shielding and heat removal requirements, and mine loading can be increased somewhat. Further study is needed to establish an optimum age for waste burial, but this age is likely to be at least ten years.

Limiting planar heat input criteria for HLW have been established.(28) A limit of about 150 kW/acre has generally been assumed, but the design presented herein assumes a somewhat lower value (126 kW/acre). The facility is sized to receive all the HLW, the cladding wastes, and the other transuranic wastes expected to be generated by the LWR fuel cycle through the end of the century.(17,19) This corresponds to all the wastes generated by 140 GW of installed nuclear capacity over a 30-year lifetime or the equivalent of approximately 107,000 MT of heavy metal originally charged to the reactors. Thus, for the 700 GW additions to total LWR capacity assumed in this study (see Task A report, Section 5.6), five such reference facilities must eventually be built. It should be noted that the 140 GW facility size is purely arbitrary. Should the number of suitable sites available for waste burial be limited, a greater amount of waste can be placed at an individual site.

B-3.5.1.1 Waste Packaging

For purposes of this study, it has been assumed that HLW and cladding/intermediate TRU are packaged in canisters with the same dimensions in order to permit standardization of shipping casks, waste receiving and handling equipment, and in-mine waste handling and placement equipment.(17) Canister size for a given facility will depend on whether HLW or spent fuel is being buried. For HLW, a cylindrical canister with a 30.5-cm (12-in) i.d. and a length of 3.05 m (10 ft) has been suggested. Canister designs have not been developed to date for a throwaway fuel cycle, but would probably be approximately the same diameter and about 4.57 m (15 ft) in length, i.e., long enough to contain a fuel assembly packaged intact.

It should be noted that cladding and, in general, the TRU wastes produce only small quantities of heat.(17,19) Furthermore, TRU will require less shielding than HLW. It is, therefore, believed that the use of shielded bulk shipping containers will be considered for TRU. Such containers could consist of stainless steel-lined, pre-fabricated concrete of sufficient thickness to provide shielding that will permit direct personnel contact and handling with conventional equipment, such as fork lifts.

Packaging of low-level TRU wastes in standard 208-liter (55-gallon) drums with a unit package consisting of 12 such drums has been suggested.(17) Bulk packaging in 1.4-m^3 (50-ft^3), 2.8-m^3 (100-ft^3), or 5.6-m^3 (200-ft^3) containers should also be considered to optimize handling and costs.

B-3.5.1.2 Waste Receipt at Facility

The waste burial facility must be capable of receiving shipments of all forms of waste by either truck or rail transport. Shipments of low-level TRU will probably be made by truck, with shipment of up to 8 units of 12 standard 208-liter (55-gallon) drums each, or an equivalent freight volume of bulk shipping containers. Canistered waste shipments of HLW, spent fuel, cladding hulls, and intermediate-level TRU will probably be made by rail in large shielded casks capable of transporting approximately 10 to 12 canisters each. Receipts will be assumed to occur over a ten-year period.

A cask is a massive shielded, sealed container, with heat removal capability, designed to transport radioactive material safely through non-restricted areas. The cask must be designed to have a very low probability of radioactivity release, even in the event of a serious shipping accident. Such rail casks (for HLW canisters) have not yet been developed, although the technology is readily available from experience with spent fuel casks. Limiting cask design parameters are expected to be in the areas of shielding, heat removal systems, weight, and size. Cask designs must be reviewed and licensed by the U.S. Department of Transportation prior to use. Because of weight limitations on highway transport, smaller casks designed for truck shipment will be able to carry only one or two canisters per shipment.

The amounts of the various materials to be received at the reference facility are shown in Table B-12. Since a total of approximately 350,000 canisters must be received at the HLW facility, and as many as 450,000 canisters at a throwaway cycle facility, the larger volume-per-shipment rail shipping mode is indicated. If bulk shipment of intermediate-level TRU and/or cladding hulls is shown to be feasible, the shipments may be made by truck rather than rail because of the greater flexibility in handling and routing.

B-3.5.1.3 Surface Facility

The surface facility must include all the buildings and services required to support the receipt, handling, and in-mine placement of the various types of waste. Essential components of the surface facility are discussed briefly in Appendix B-IV. The equipment, safety systems, and building structures must be designed to appropriate codes and specifications in order to ensure the health and safety of the operators and the general public during both normal operations and abnormal occurrences. NRC has yet to publish specific design criteria for waste burial facility construction and operation.

B-3.5.1.4 Site Access

A rail spur and access highway must be extended to the repository site. Adequate switching facilities and rail siding must be provided on-site in order to receive and handle efficiently as many as 10 to 12 rail casks per day for the receipt of HLW spent fuel, cladding hulls, and intermediate-level TRU canisters.

Electrical, sewer, and water services must also be extended to the site.

TABLE B-12

VOLUMES OF WASTE TO BE STORED IN REFERENCE FACILITY

Type of Waste	Unit Packaging(*) Description	HLW Facility(*)		Throwaway Facility
		No. of Waste Units	Volume (m^3)	No. of Waste Units
HLW	canister[+]	35,500	10,750	319,500
Cladding hulls	canister[+]	58,500	9,240	0
Intermediate-level TRU	canister[+]	255,000	46,800	130,000
Low-level TRU	55-gal. drum	655,000	140,000	330,000

*Based on information contained in ERDA-76-43, Appendix C.

[+] HLW canister is 30.5 cm diameter x 3.05 m long, total weight is about 900 kg. Spent-fuel canister would be about the same diameter and 4.6 m long; total weight would be about 1200 kg.

B-3.5.1.5 Below-Ground Layout

B-3.5.1.5.1 Thermal Considerations

In order to minimize area commitments and waste disposal costs, it is desirable that a maximum amount of HLW be placed in a given mined area, subject only to as yet imperfectly-understood constaints on temperature distributions in the repository. Certain trade-offs must be evaluated, using good engineering judgment. The major areas to be considered in setting limits on repository temperature distributions are as follows:(2,28) These should be viewed only as possible considerations relating to repository thermal loading.

- Maximum allowable temperature at the centerline of the waste container

  For glass, the centerline temperature limit is considered to be on the order of 600°C, somewhat below the devitrification level for glasses and substantially below processing temperatures in order to preclude additional off-gassing.

- Temperature limitations immediately adjacent to the storage canister

  It has been suggested(2,28) that temperatures of 250°C should not be exceeded in more than about 1% of the salt within the boundaries of the entire canister array. This temperature limit is set by concern over too high a surface temperature near the canisters. The corrosion problems associated with it would also minimize the amount of salt in a highly plastic state in the immediate canister region, i.e., extending about 20 cm into the salt from the canister surface.

- Temperature limitations throughout the mined area

  Because of the salt plasticity, temperatures must be limited to values that will not cause stability problems during mining operations. Somewhat higher bulk temperatures are desirable after mine closure, however, in order to insure fairly rapid filling of voids. Taking account of both considerations, it has been estimated that about 200°C should be used as a temperature limit for approximately 25% of the salt volume. In the analysis of salt repository layouts this limit has been equivalent to constraining the peak temperature at the midpoint between canister positions. The 200°C temperature limit is actually set by a plastic flow analysis in which the calculated gross thermal expansion of the salt field area would be offset by "collapsing" of the void volume in the salt cavity itself. The specifics of this balance will differ among designs and the calculations themselves are imprecise. This particular thermal limit value may well be modified as salt mechanics become better understood.

- Temperature rise in fresh water aquifers

  Fresh water aquifers may be adjacent to or above the repository. It is desirable that the temperature of these aquifers not rise significantly; temperature rises of about 10°C at a 30-m depth and 30°C at a 90-m depth would be "acceptable."(28) It appears that these values are achievable.

- Stability of the geologic formation when subjected to stresses caused by thermal expansion

  Overall net surface expansion should not be so great as to create geologic stresses that would cause a breach of repository integrity. This concept is very important, because it directly affects the planar heat density and, hence, mine size. More effort is needed to determine the importance of such a limit and to set appropriate values.

- Ground surface temperatures

  It has been suggested that the average surface temperature of the ground not exceed 0.6°C (1°F) above the ambient.(28) This is easily achieved.

- Temperatures beyond the property boundary

  Beyond the buffer zone, subterranean temperatures should be limited to a 0.6°C rise.(28) The value appears to be quite restrictive, requiring a fairly large exclusion radius; furthermore, the rationale for choosing this low value does not seem to have been developed in the literature.

When all of these factors are considered together, they result in a planar heat input limit (for a salt repository) of about 150 kW/acre for ten-year old HLW.(28) For HLW of a younger age, the value would be somewhat greater at the time of deposit, while for an older waste, the value would be reduced appropriately. If the wastes are buried earlier, the net effect would be to increase repository size. This increase relates primarily to the "gross salt temperature limit," e.g., that 25% of the total volume not exceed 200°C. The local temperature effects, e.g., the canister centerline temperature, or the 1% salt criterion, can be met simply by different packaging approaches, such as use of a smaller-diameter canister or a less dense heat source.

The value of 150 kW/acre was developed several years ago for a salt bed approximately 305 m (1000 ft) below the surface. More recent studies have generally continued to assume the same or somewhat lower planar heat densities. Because an independent repository design was beyond the scope of the present study, the latest OWI design(17) has been applied to the 460-m (1500-ft) repository depth. This design has a planar heat density of about 126 kW/acre. If bulk salt mine temperature is limiting, the planar heat density limit is relatively insensitive to repository depth because of the very low rate of heat transfer to the surface. Heat storage dwarfs heat transfer from the repository for several hundred years. Hence, changes in depth, which affect the rate of heat transfer, will have only a small effect on peak salt mine temperatures.

If uplift caused by thermal expansion should turn out to be the limiting thermal criterion, planar heat density limits will be somewhat more sensitive to depth, since the amount of thermal expansion depends upon the amount of total stored heat within the geologic formation. For a depth of 460 m, a maximum stored heat condition is reached between 500 and 1000 years. Maximum uplift forces caused by thermal expansion will be reached at the same time.

Based on calculations, the ratio of total stored heat for spent fuel vs HLW is in the range of 2-2.5 during the first 1000 years. The heat production rate for spent fuel is higher than for HLW over extended periods because of the larger quantities of long-lived transuranic isotopes present in spent fuel. If it is assumed that the total stored heat and the resulting physical impact on the geologic formation are limiting, then the planar heat density for a spent fuel burial field must be reduced accordingly. In order to estimate a reference mine layout and waste disposal facility costs (see Section 4.0), it has been assumed for illustration that the planar heat density for a spent fuel burial facility will be approximately 40% of that for a HLW facility. Therefore, 2.5 times the total mine area needed for HLW would be necessary to store an equivalent amount of spent fuel wastes. Investigations have not revealed any specific criteria for limits on thermal expansion; it is possible that greater thermal expansion than has been assumed in the OWI reference design may prove acceptable.

B-3.5.1.5.2 Mine Level General Arrangement

The mine level will consist of segregated storage areas for HLW, spent fuel, cladding hulls, and TRU, a distribution system for ventilation intake air, a network of ventilation exhaust tunnels, and mine level support facilities. (A detailed description of the mine facility will be found in Appendix B-IV; a conceptual sketch is shown in Figure B-18.) The mine design will be in accordance with Federal health and safety standards, "Metal and Non-Metallic Underground Mines," 30 CFR 57, and other appropriate mining codes and practices. The design must meet requirements for ventilation, mine opening and mine cavity stability, the effects of waste heat input and salt heat transfer coefficients on facility stability, the efficient use of mining and hauling equipment during mine construction, and the efficient placement of waste during mine loading.

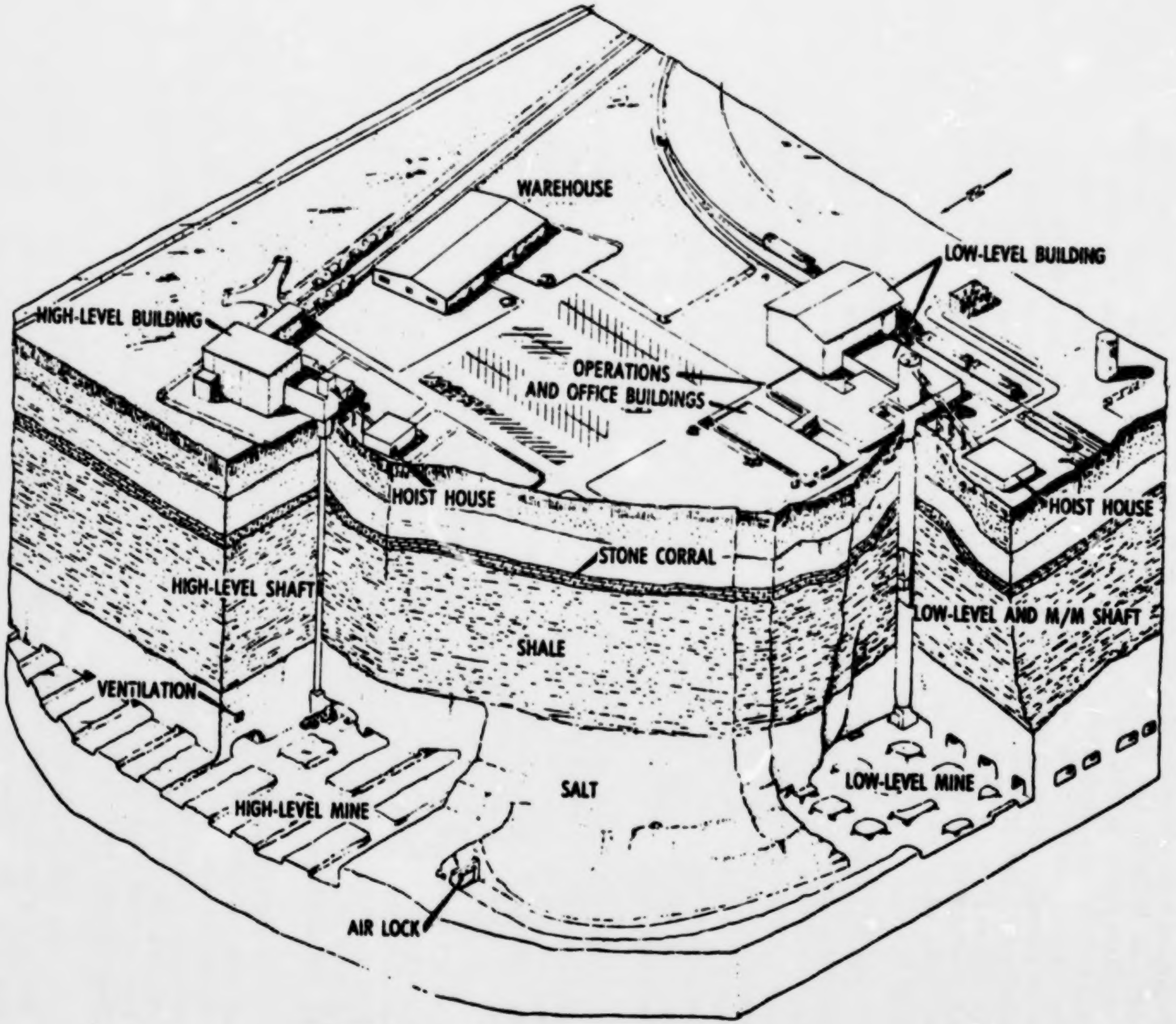

WAREHOUSE
LOW-LEVEL BUILDING
HIGH-LEVEL BUILDING
OPERATIONS
AND OFFICE BUILDINGS
HOIST HOUSE
HOIST HOUSE
STONE CORRAL
HIGH-LEVEL SHAFT
LOW-LEVEL AND M/M SHAFT
SHALE
VENTILATION
SALT
LOW-LEVEL MINE
HIGH-LEVEL MINE
AIR LOCK

FIGURE B–18 ARTIST'S CONCEPT – FEDERAL REPOSITORY

Source: Zerby, C.D. The National Waste Terminal Storage Program. Proceedings of Symposium on Waste Management, Tucson, Arizona, October 1976, CONF 761020.

Once a maximum permissible planar heat density has been determined, based on heat transfer calculations, the waste canister disposal array and canister-to-canister spacing can be established. There must be a compromise between the most uniform heat transfer array (uniform spacing of canisters throughout the unit burial volume) on the one hand, and mine stability, mining economics, and ease of waste placement on the other. These latter considerations require that the waste be placed in long narrow tunnels. Heat transfer calculations must again be made for the center-to-center spacing chosen in order to verify that the limiting salt temperature criteria (Section 3.5.1.5.1) are still met. The total heat loading per acre that is possible in the tunnel array may be slightly less than that for a uniform canister array because of localized heating effects.

For this study,HLW canisters are assumed to contain the wastes from 3 MTHM charged as fresh fuel to the nuclear reactors. For spent fuel, nine canisters will be necessary to bury this equivalent amount of waste, based on the assumptions that each canister contains one spent fuel assembly. For a planar heat density that is 40% of that for HLW, approximately 3.6 times as many spent fuel canisters will need to be buried per acre. Loading spent fuel canisters with two or more fuel assemblies per canister may be possible, thus reducing canister handling operations and final burial costs.

Adjacent burial tunnels run parallel to each other, with rows of burial tunnels connected by a network of branch and main tunnels leading back to the canister waste receiving station/transfer hot cell at the base of the canister hoist. Mine capacity is increased by adding additional rows of burial tunnels. Tunnel and corridor dimensions are set by the size of the waste canister placement equipment, with clearances reduced to a minimum in order to minimize excavation costs. Corridors, burial tunnels, and the network of ventilation exhaust shafts are excavated using conventional mining equipment. The mined salt is removed from the corridors and burial tunnels using conventional mine materials hauling equipment that deposits the salt at the main shaft for transport to the surface.

A reference bedded-salt mine general arrangement for storage/burial of HLW, cladding hulls, and TRU has been developed.(17) Based on a review of the heat generation rate of ten-year-old HLW and on the canister handling and placement operations, it appears that the canister spacing and mine general arrangement given in Table B-13 is typical of the layout that will eventually be used in commercial waste burial facilities. This arrangement is assumed for HLW in this study as well.

The reference facility for spent fuel disposal will be similar in design, with dimensions also summarized in Table B-13. Major differences are that the tunnel and corridor heights must be raised approximately 1.5 m (5 ft) to accommodate equipment for handling the larger waste canisters; individual canister holes will hold four spent fuel canisters vs one HLW canister (the alternative is four times as many holes per tunnel);

TABLE B-13

HLW/SPENT FUEL RETRIEVABLE STORAGE AREA DATA

	HLW	Spent Fuel
Capacity (canisters)	35,500	319,500
Total MTHM charged (to reactor)	1.07 x 10^5MTHM	1.07 x 10^5MTHM
Specific heat Input	150 kW/acre (Max) 126 kW/acre (most likely)	60 kW/acre 51 kW/acre
Canister density	32 canisters/acre	115 canisters/acre
Approximate canister dimensions	0.32 m o.d. x 3.0 m long	0.32 m o.d. x 4.6 m long
Canister hole dimensions	0.46 m i.d. x 6.1 m long	0.46 m i.d. x 23.2 m long
Number of canisters per hole	1	4
Corridor dimensions	9.1 m wide x 5.5 m high	9.1 m wide x 7.0 m high
Burial tunnel dimensions	171 m long x 5.5 m wide x 5.5 m high	171 m long x 5.5 m wide x 7.0 m high
Canister center-to-center spacing	5.3 m	5.3 m
Number of canisters per burial tunnel	32	115
Number of burial tunnels	1112	2786
Burial tunnel center-to-center spacing	23.8 m	23.8 m
Salt pillar thickness between tunnels	18.3 m	18.3 m
Salt pillar thickness between burial tunnels end to end	18.3 m	18.3 m

the spent fuel storage area will have 2.5 times as many burial tunnels to accommodate the equivalent amount of waste; the cladding portion of the TRU burial area will be eliminated; and the volume of the TRU burial area will be reduced in size, because this type of waste will be drastically reduced in volume.

Dimensions for the TRU areas are shown in Table B-14. While not optimal, they are sufficient for the present.

B-3.5.1.6 Total Reference Facility Area Estimates

Table B-15 shows the area breakdown for the two reference facilities cited in this study. The total controlled area around the surface facility is assumed to be a distance of one mile radially outward from the outer edge of all waste burial fields.(28) This distance was arbitrarily set to ensure that the thermal effects at any point outside the controlled area, either on the surface or underground adjacent to the buried waste, do not result in a temperature rise of more than 0.6°C. Based on a preliminary heat transfer analysis, this would require a distance of approximately one mile from the edge of the burial field.

B-3.5.1.7 Summary of Major Differences Between the HLW and Spent Fuel Burial Facility Reference Designs

The general equipment design, surface facility structures, and mine layout concepts are similar. Major differences are in dimensions and in the number of waste units that must be handled. These differences are summarized below:

(1) Planar heat density. Spent fuel will contain more actinides (see Task A report), leading to a greater integrated heat source over the stored life of the waste. Thus, the initial mine loading must be reduced; a reduction factor of 2.5 (i.e., from 126 kW/acre for HLW to 51 kW/acre for spent fuel) would allow for the added heat production over the first 1000 years.

(2) Canister sizings. HLW canisters will be 3 m (10 ft) long, and spent fuel canisters will be approximately 4.6 m (15 ft) long. This extra length will change the size of shipping casks, surface handling facilities, mine facilities, and mine handling equipment.

(3) Excavation requirements. More than three times as much material must be mined for a spent fuel disposal facility as for a HLW facility.

(4) Number of canisters. As many as nine spent fuel canisters must be buried to account for the equivalent amount of waste in a HLW canister. Burial of four spent fuel canisters per

TABLE B-14

REFERENCE FACILITY DIMENSIONS FOR TRU WASTE

Cladding Hull/Intermediate-level TRU Burial Field Data

	HLW Facility[*]	Spent Fuel Facility
Waste container	canister	canister
Canister dimensions	0.3 m i.d. x 3.0 m long	0.3 m i.d. x 4.6 m long
Capacity - cladding hull canisters	58,500	0
Capacity - TRU canisters	255,000	less than 130,000[+]
Burial room dimensions	171 m long x 11.6 m wide x 5.5 m high	171 m long x 11.6 m wide x 7.0 m high
Number of canisters per room	790	790
Canister spacing	6 rows, 1.2 m center to center	6 rows, 1.2 m center to center
Number of canisters per hole[‡]	1	1
Spacing between burial rooms	23.8 m center to center	23.8 m center to center
Number of burial rooms	398	165

Low-Level TRU Burial Field Data

	HLW Facility[*]	Spent Fuel Facility
Waste container[‡]	55 gal drum	55 gal drum
Capacity, TRU drums	655,000	less than 330,000[+]
Burial room dimensions	171 m long x 11.6 m wide x 5.5 m high	171 m long x 11.6 m wide x 5.5 m high
Type of storage	Warehouse	Warehouse
Number of drums per room[+]	7,000	7,000
Number of rooms	94	47

---

[*] Source: Parsons, Brinckerhoff, Quade & Douglas, Inc., Waste Isolation Facility Description, Y/OWI/SUB-76/16506, Office of Waste Isolation, 1976.

[+] Preliminary estimate.

[‡] May not represent optimized design.

TABLE B-15

TOTAL FACILITY AREA REQUIREMENTS

	HLW Facility (Acres)	Spent Fuel Facility (Acres)
Total controlled area	10,600	12,000
Total area containing waste*	2,000	3,487
HLW/spent fuel	1,112	2,786
Cladding hulls/Intermediate-level TRU	398	165
Low-level TRU	94	47
Mine shaft exclusion area, corridors service area, etc.	400	500

*Includes the area of tunnels plus area between tunnels.

hole can reduce the number of holes to 2.5 times the number required for HLW.

(5) Number of canisters per unit burial tunnel. As many as 3.6 times as many spent fuel canisters (9/2.5) as HLW canisters will be buried in each unit burial tunnel. This can be accomplished either by 1) boring 3.6 times as many canister holes (probably resulting in the need for wider tunnels); 2) multiple canister loadings in the same number of canister holes (drilled deeper) as used for HLW; or 3) some combination of each.

(6) Impact of TRU wastes. For the throwaway cycle, much less TRU waste is generated, reducing the mine volume needed. The reduction is offset to some extent by the greater number of spent fuel canisters than HLW canisters.

B-3.5.1.8 Assessment of the State of the Art for Burial in Salt

Heat transfer characteristics and the behavior of salt at elevated temperatures can be readily predicted. Within the thermal criteria chosen (no more than 1% of the salt over 250°C, and no more than 25% over 200°C), general mine stability remains high and local heating around the canister permits enough plastic salt flow to seal the canister in the salt formation. Much practical experience has been gained in the Project Salt Vault Studies.(29)

Mine construction and handling equipment needs appear to be well within current technology and should prove to be straightforward engineering jobs. Salt mining can be accomplished using conventional mining techniques. Canister-handling equipment must perform relatively simple, straightforward operations, and although all handling must be done by remote control, equipment design should be well within the current capabilities of the nuclear industry.

B-3.5.1.9 Facility Design Refinements

The reference facilities in this study represent typical arrangements, but it is not suggested that they represent an optimized design in every respect. Several areas where further studies may lead to a less costly facility are suggested below:

- Optimize age of waste to permit balancing higher mine-loading density against cost of longer-term surface storage.
- Consider loading low-level TRU in HLW tunnels before backfilling the tunnels, eliminating the need for a low-level TRU storage field.

- Consider the use of bulk shipping containers for low-level TRU, as alternatives to drums.

- Evaluate the use of self-shielded intermediate-level TRU bulk shipping containers instead of canisters. Bulk containers could be handled directly, using conventional freight-handling equipment, warehouse-style, in the mine. The number of containers to be handled can be reduced by a factor of 10.

- Evaluate the possibility of loading more than one spent fuel assembly in each canister to decrease the number of holes that must be bored in the burial tunnels.

B-3.5.1.10 Retrievable Storage

The reference facilities discussed to this point have assumed permanent, non-retrievable waste storage. Under certain circumstances, however, it may be desirable to store the wastes in an arrangement that is suitable for permanent disposal, while retaining the option to retrieve the wastes within a predetermined period of time. Retrievability might be useful for the pilot waste burial facility, in order to facilitate periodic canister inspection and testing. Retrievability may also be useful for spent fuel burial, although this might better be done in near-surface facilities. Although the present policy on reprocessing of spent fuel is unsettled , it seems unlikely to many people that spent fuel, with its intrinsic value as an energy resource, will actually be "thrown away", but rather that spent fuel would be stored in a facility from which it might be recovered and reprocessed at a later date.

The general arrangement for a retrievable deep storage facility would probably be similar to that for non-retrievable deep storage in many respects. The surface facility and the mine access shafts perform the same functions and will be identical. The general mine layout will also be similar, with canisters of waste buried in holes in the floor of long narrow burial tunnels. Mine stability and integrity over the full length of the storage period must be verified prior to setting the canister spacing and tunnel dimensions for the particular geologic medium, but spacing and dimensions are expected to be similar for both retrievable and non-retrievable storage.

There may be design differences in the following areas:

- Mine Ventilation - The system for a retrievable facility may have to be designed for a higher capacity, because a larger unfilled mine volume must be ventilated over a much longer period of time. The ventilation system may also be used to remove decay heat from the mine over the retrievable period as a measure to further enhance mine stability.

- Mined Materials Removal System - Much greater quantities of mined salt must be removed from the mine and stored on the surface for a retrievable facility. For non-retrievable storage, once a working mine volume has been created, salt from a tunnel under construction is used as backfill for a tunnel already loaded with waste. In this way, the total volume of salt transferred to the surface is minimized. For retrievable facilities, all the excavated material must be transferred to the surface, stored for the desired period of waste retrievability, and then returned to the mine as backfill material during the facility decommissioning period. For the reference facilities assumed for this study, approximately 37 x 10^6 m^3 must be removed from a retrievable spent fuel burial facility while as little as 2.5 x 10^3 m^3 must be removed from an equivalent permanent spent fuel burial facility.

- Canister Burial Hole Arrangement - For non-retrievable storage, a canister will simply be placed in a hole bored in the floor of a waste burial tunnel and the hole will be backfilled up to floor level with crushed salt to provide radiation shielding to the operators. To facilitate retrievability, however, the canister hole design must be changed. The canister hole will probably have to be lined to prevent plastic flow in the salt from sealing the canister in the salt, and have to be sealed with a concrete plug to provide shielding. Carbon steel 25 cm (1 in) thick is suggested as a liner material in order to maintain enough integrity over the desired period to prevent the surrounding salt from collapsing around the canister.(17) A minimum of five to six feet of salt thickness will probably be required to provide adequate shielding (less than 10 mR/hr) at the surface of the tunnel floor for 10-year old wastes.

- Waste Placement Equipment - Additional equipment, such as a shielded canister hole isolation valve and a canister hole plug installation machine may have to be developed to permit waste placement in retrivable storage.

- Canister Material - Retrievability may require that the canister material be upgraded to one with long-term corrosion resistance (perhaps titanium).

- Facility Decommissioning - For non-retrievable storage, tunnels are backfilled with mined materials from other burial tunnels under construction. Thus, ultimate facility decommissioning consists simply of surface facility decommissioning, with contaminated materials from the surface facility placed in open spaces, such as mine corridors, followed by backfilling of the mine and, finally, of the mine shaft, as determined by regulatory policy at the time of decommissioning. Decommissioning of a retrievable facility will be a more costly operation.

B-3.5.2 Disposal in Other Media

B-3.5.2.1 Summary Discussion of Thermal Criteria

To date, most of the detailed design work on geologic disposal facilities has been limited to salt deposits, but alternative geologic media are of interest for the reasons discussed at the beginning of Section 3.5. Crystalline rocks, such as basalt or granite, appear to be of greatest interest, although consideration is also being given to shales and other sedimentary rocks.

Because of lack of detailed engineering data, it has not been possible to perform a thorough assessment of disposal technology in media other than salt. This section will, therefore, discuss only briefly certain aspects of facility design that might have a bearing on the integrity of a repository in such media as granite or basalt. Some limited engineering data are available for crystalline rock repositories, but there seem to be essentially no data on other rock types, such as shale, and therefore they will not be considered further.

The importance of thermal criteria has been pointed out in the review of salt repository designs and, clearly, they will also have a significant impact on facility design in all geologic media. For this reason, scoping analyses of thermal characteristics have been performed for granite as well as for salt (Section 3.5.3). In reviewing thermal design criteria, one of the first questions that arises is whether there are analogies between the "defined" criteria for salt and the potential criteria for granite. A comparison of the two media is given below.

- Temperature limitations immediately adjacent to the storage canister. The limit could possibly be higher for granite than for salt, since corrosion should be much less of a problem in granite and because plastic flow does not occur in granite as it does in salt. On the other hand, the lower thermal conductivity of granite leads to steeper temperature gradients than in salt. Such gradients could cause localized cracking and/or spalling in the vicinity of the canister. For non-retrievable storage, a limited fracture zone should not present serious problems; this would not be the case for retrievable storage, however. In any case, the extent of such fractures could be controlled by reductions in canister size and/or heat release per canister.

- Temperature limitations throughout the mined area. The problems are of the same nature in either medium; the primary concern is stability of the repository. Granite is a less satisfactory medium in this respect, because of the absence of significant plastic flow and the associated intolerance of significant temperature gradients. To set thermal criteria, a detailed analysis of the rock mechanics is necessary. Some work on granite repositories has been reported in Canada including at least a partial analysis of rock mechanics in the mined area, indicating that structural integrity can probably be achieved at bulk temperature limits somewhat less than those developed for salt.(30)

- Stability of the geologic formation when subjected to stress caused by thermal expansion. As already noted in the discussion of salt repositories, criteria on maximum permissible uplift are highly tentative and probably subject to evolutionary changes as more work is performed on rock mechanics. The Canadian studies do not appear to have considered the detailed mechanics of the thermal expansion problem, which is very complex in any case, and especially for granite where there is little plasticity and for which the fracture mechanics are imperfectly understood. Conservative scoping calculations of thermal uplift (to be discussed below) lead to modest values of 1.2 m (4 ft) at 150 kW/acre. These scoping calculations cannot allow for the nature and extent of cracking that might result from any given pattern of uplift.

The remaining four considerations--a) canister centerline temperatures, b) temperature rise in fresh water aquifers, c) ground surface temperatures, and d) temperatures beyond the property boundary, are relatively independent of type of geologic medium.

B-3.5.2.2 Canadian Studies on Granite Repositories

Atomic Energy of Canada Limited (AECL) has been considering rock repositories for some time, and has funded research and engineering programs to calculate rock temperature distributions as well as the resulting stresses and failure mechanics associated with these temperature distributions. A recent study by Acres Consulting Services Limited (Acres) covered analysis of temperature distribution and failure modes in a granite repository designed for retrievable spent fuel storage over a time period of 30 years.(30) This study utilized a planar heat density of about 130 kW/acre keyed to a five-year-old waste. Allowing for differences in fuel characteristics and adjusting the 130 kW/acre figure for a ten-year age results in an equivalent planar heat density of 80-90 kW/acre or about 60% of the nominal level for HLW in salt (150 kW/acre). The granite depth in the Acres study was at the 1000-m level (more than 3000 ft) compared with the reference depth in this report of 460 m (1500 ft). At a 1000-m depth, the resulting allowable planar heat density is less because of greater stored energy in the repository prior to reaching equilibrium heat-transfer conditions. (At greater depths, longer times are required to reach the peak temperature condition.)

For the stress analysis, a two-dimensional finite-element computer program was used for steady-state and transient calculations. In carrying out these calculations, certain assumptions had to be made regarding the (a) rock joints and their strength; (b) mechanism and criteria for failure; (c) non-linear rock behavior under long-term mechanical loads; (d) stabilization pressures as related to long-term stability; (e) influence of pre-existing rock stress. These assumptions were necessary because of inadequate physical data and/or model limitations. To compensate for uncertainties, the investigators used conservative input parameters.

Temperature and stress contours were developed and resultant failures were presented in a series of sectional views. This paper concluded that although minor failure occurred at the bottom and top corners of the repository tunnels, these failures were not likely to propagate far into the rock, and the basic integrity of the whole structure would probably be maintained. On the other hand, anchors might be required to prevent collapses of the top wall in certain areas if it were important to minimize local build-up of rubble. The degree of failure during the first 30 years would be to a very large extent influenced by the amount of ventilation and cooling that occurred during this period,which is characterized by high thermal gradients.

The foregoing remarks applied to the mine structure above the tunnel floor. For the floor area containing the waste canisters, it seems probable that cracking would occur both adjacent to the canister and above it. Such cracking could make subsequent retrieval or removal of the canister difficult. It was therefore tentatively concluded that for a retrievable facility these holes would have to be lined.

The conclusions of the study were summarized by the authors:(30)

(1) Failure of the jointed rock mass around a ventilated room is restricted to the portions of the room below the springline.* The requirements for supporting these regions are minimal.

(2) The floor area needs further engineering to ensure retrievability of the waste canisters. Detailed analyses of the stresses and the displacements in this region should be performed using a discontinuum model and field data (heater experiments, geotechnical investigations).

Although much more analysis and information are needed to design a granite repository, the Acres study suggests that such a repository is feasible.

B-3.5.2.3 Possible Thermal Criteria for Granite

In any given medium, temperatures in the immediate vicinity of the canister are controlled mainly by canister size and heat output. If localized cracking is deemed to be a problem for non-retrievable storage, it can be controlled by using smaller-diameter canisters and/or lower heat release per linear foot of canister. For the same planar heat density, lower canister heat output would necessitate closer spacing, with more holes and therefore higher costs. Presently, such parameters cannot be estimated with any precision because of the lack of detailed data on localized cracking from high thermal gradients in granite. For the purpose of this study, there seem to be no inherent feasibility problems with further subdivision of wastes and/or lining of waste holes if this should prove necessary.

---

*The line of intersection of roof and walls.

As for temperature limits in the mine itself, it would appear from the Acres study that a modest reduction from the planar heat density assumed for salt in this report would possibly be sufficient to assure satisfactory integrity of the mine area proper. The planar heat density also affects gross thermal expansion and uplift of the repository area, however. The thermal analysis presented below shows that temperature distribution outside the repository does not differ greatly between salt beds and granite. These temperature distributions are important in determining the thermal expansion characteristics of the repository.

It is possible that planar heat density may be set by limitations on thermal expansion rather than on mine temperature. Vertical thermal expansion, i.e., the rise in the surface ground elevation compared with its original level, may be especially important if it results in connected fractures of the granite. Table B-16 provides perspective on these effects, based on a very simple expansion model. This table is based on a planar heat density of 150 kW/acre. The indicated expansions are gross expansions and any subsidence (collapse of the mine void area) would have to be subtracted from the gross expansion figures.

The reference salt bed repository indicates a gross expansion of 2.8 m. This value conservatively assumes that the three-dimensional volumetric expansion characteristics of the salt and the overburden all act in the vertical direction and therefore the vertical expansion coefficient is effectively three times the linear expansion rate. This is perhaps not an unreasonable assumption for salt because of its high plasticity. It is probably an overly conservative assumption for a crystalline rock where horizontal expansion may not be converted to vertical expansion unless there are suitable slip planes or connected fractures within the gross rock structure. Nevertheless, the table indicates that the granite repository would have an expansion approximately one-third that of the bedded salt.

In comparing the calculated thermal uplift data with other published results, there is reasonable agreement when corrections are made for differences in depth and planar heat densities. For example, the Culler Report indicates a thermal expansion of about 1.5-1.8 m (5-6 ft) and a subsidence of about 1.2 m (4 ft), with these phenomena occurring about 200-300 years from deposition.(31) The calculated value is higher and occurs somewhat later in time because of the greater depth of the reference repository, but it is consistent with the Culler Report data. This good agreement lends confidence to the preliminary calculations for granite repositories.

TABLE B-16

CONSERVATIVE ESTIMATES OF GROSS VERTICAL EXPANSION ("THERMAL UPLIFT") FOR DIFFERENT REPOSITORY MEDIA

	Time of Maximum Stored Heat	Expansion[*]
Salt bed	1000 years	2.8 meters
Granite	1000	1.2
Salt Dome	500	7.6

---

[*] Assumes that all of volume expansion is in vertical direction (3 times linear expansion coefficient), i.e. an ideally plastic material. Linear coefficient assumed to be 40 x 10^{-6}/°C for salt and 7.9 x 10^{-6}/°C for rock.

An arbitrary limit on planar heat density one-half that of the corresponding values for salt, should be a sufficiently conservative assumption to allow for uncertainties in characterizing granite repositories. Uplift would be calculated to be less than 0.6 m (2 ft) and peak mine temperatures would be significantly less than those calculated in the Acres study. It is, therefore, an appropriate assumption for the purposes of this study, although it should be considered only as a "best-guess" at this time, recognizing current state-of-the-art limitations on design of repositories in granite.

B-3.5.3 Thermal Analysis

B-3.5.3.1 Background

Thermal design criteria for a HLW repository have a major impact on the size and geometry of such facilities. Suggested thermal design(28) criteria for a "bedded salt" repository at a depth of approximately 305 m (1000 ft) have been published and are summarized in the discussion of the reference salt repository design (Section 3.5.1).

It is beyond the scope of this study to perform a detailed thermal design of a repository; nevertheless, some independent calculations have been made in order to demonstrate the effects of variables such as time, medium, waste age, planar heat density, and waste type. Broad conclusions may then be drawn regarding the effects of different waste management plans and standards on repository design.

Although no criteria have been established for the design of a rock repository facility, an understanding of the temperatures involved can give some insight as to what densities of storage might be attainable.

Three different types of thermal analyses have been performed, each of which fills a specific need. These are:

1. A three-dimensional (3-D) analysis run in order to determine the short-term temperature behavior near the waste containers. This analysis is necessary to determine if the "1%-salt-at-250°C" criterion is met. It is at this time that maximum thermal gradients exist.

2. A one-dimensional (1-D) analysis run in order to determine the long-term temperature distribution gradients in the vertical direction, both above and below the repository area. It is from this temperature distribution that the maximum integral energy deposit can be calculated and from which the maximum possible thermal expansion can be estimated. This temperature distribution is a function of the type of rock as well as of the depth of the repository. If uplift from thermal expansion should become a limiting factor, planar heat density would be set by this temperature distribution.

3. A two-dimensional (2-D) analysis run in order to determine the added heat transfer that will occur from the repository area by virtue of radial heat flow and to establish if this effect has much of a bearing on the temperature at the vertical centerline of the facility. This analysis is also useful in determining the temperature distribution beyond the facility itself in order to set buffer zone distances.

The initial planar heat density (kW/acre) is treated as a parameter in the calculations as is the age of the wastes at time of deposit.

Calculations have been performed for three alternative repository concepts: a bedded salt reference case, a salt dome case (similar to bedded salt except that the salt zone is much thicker in the vertical direction), and a granite repository. All three cases assume the HLW deposit to be at the 460-m (1500-ft) level.

B-3.5.3.2 Summary of Results

The results of the analyses are presented in a series of graphs that display the temperature distributions for various points in time and space, and for the assumed variation in input parameters (planar heat density and age of wastes). The analytical model and calculational results are described in detail in the remainder of this section, but a few general comments are in order:

1. Considering the repository as a whole, the temperature distributions developed for a given planar heat density do not differ greatly for bedded salt, salt domes, or granite media. This may seem surprising, since salt has generally been thought to be superior because of its higher thermal conductivity; the lower conductivity of rock is compensated for, however, by a significantly higher volumetric heat capacity.

2. The local peak temperature in granite for a given initial planar heat density (e.g., 150 kW/acre) and canister energy (e.g., 3.95 kW/canister) would be substantially greater than in salt. This may necessitate the burial of less waste per canister, but with a closer canister-to-canister spacing for granite.

3. The temperature distributions as developed for spent fuel disposal are not dissimilar to those of HLW. This is because later, when the higher long-term energy production from spent fuel is present, the heat transfer to the atmospheric heat sink has already started to dissipate the stored energy. This is the case for a 460 m (1500 ft) repository level; it may not be so for much deeper repositories.

4. A one-dimensional analysis on the vertical centerline of the field is a very good representation of the temperature distribution and two-dimensional heat transfer becomes important only over very long time frames, i.e., longer than 10,000 years.

5. The peak temperature rise in the mine is roughly proportional to the original planar heat density. For example, with ten-year-old waste at 150 kW/acre the maximum temperature rise in salt is approximately 260°C, but in the same repository at a density of 60 kW/acre (40%), the temperature rise is 90°C (35%).

6. The calculated temperature distributions in bedded salt are in good agreement with those from other studies.(28,32,33)

B-3.5.3.3 Modeling of the Repository

For these calculations, in accordance with the reference design, the waste repository was assumed to be 460 m (1500 ft) below the ground surface. The canister spacing was 23.8 m (78 ft) apart in the X and 4.3 m (14 ft) in the Y direction in a rectangular array. Figure B-19 is an isometric sketch of the layout used in the 3-D computer simulation. Since a rectangular coordinate system is used in the calculational model, the cylindrical waste container was simulated by a container having a square cross section of the same size as the cylindrical container, therefore, the surface area of the container is increased by about 13%. For the 1-D analysis the heat source was assumed to be homogeneously distributed in a 30.5-m (100-ft) thick layer of salt or granite. For the 2-D analysis the waste field was assumed to be a "pancake" 30.5 m (100 ft) thick and having a diameter of 1190 m (3900 ft).

The values of the thermal properties for salt, shale, granite, and sedimentary deposits used in the analysis are given in Table B-17.(28,32,33) The thermal conductivity of salt and granite, and the specific heat of granite were assumed to be a function of temperature as shown in Figure B-20. All other properties were assumed to be constant with temperature.

The computer code HEATING-3 was used to perform the 3-D, 2-D, and 1-D analyses.(34,35) To reduce computer running time, the explicit solution method was utilized.(36) The time-dependent heat generation rates, as used for the analysis, are given in the Task A report for the throwaway and the mixed-oxide-recycle fuel cases respectively.

B-3.5.3.3.1 Results of the 1-D Analysis

The objective of these computer runs was to analyze the long-term temperature behavior of the geologic formation above and below the mine containing the waste canisters. Three different geologic formations were considered, as given in Table B-18.

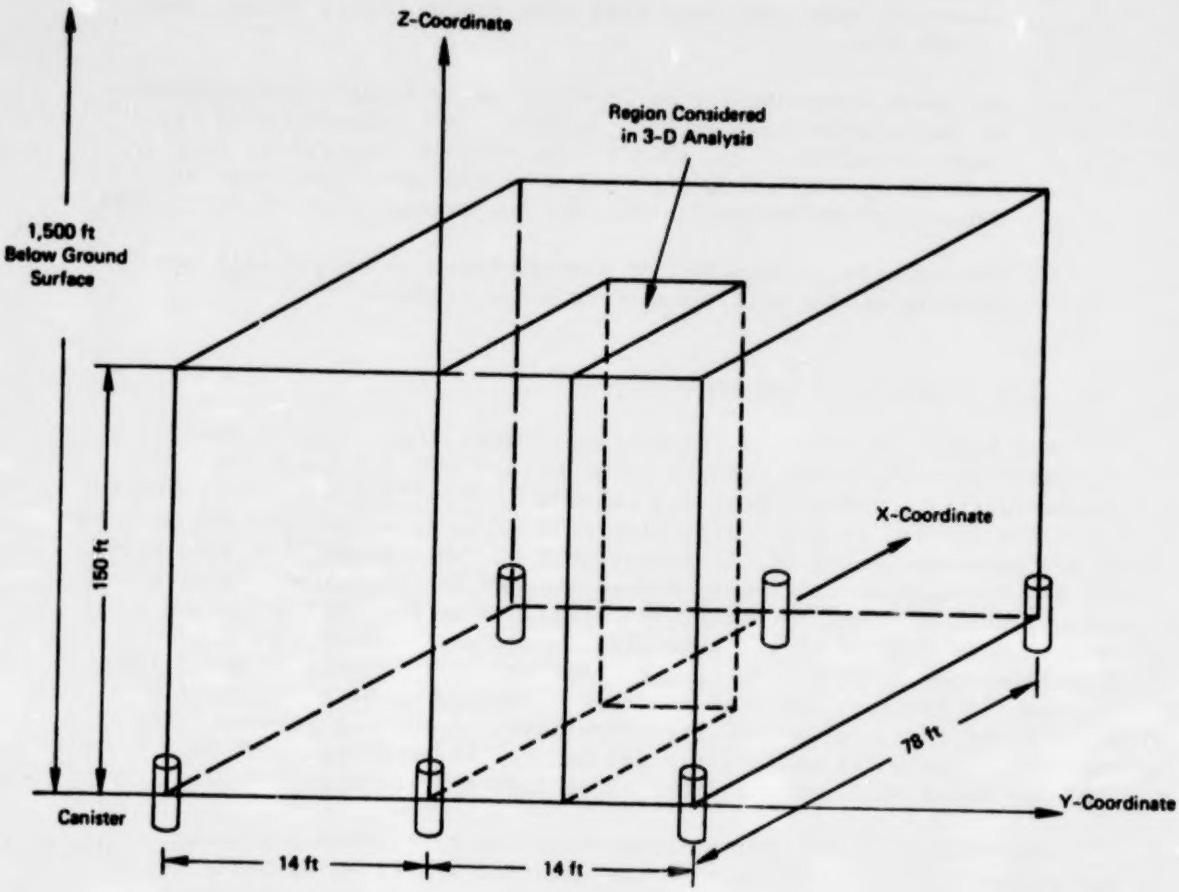

Z-Coordinate
Region Considered in 3-D Analysis
1,500 ft Below Ground Surface
150 ft
X-Coordinate
78 ft
Y-Coordinate
Canister
14 ft
14 ft

FIGURE B–19 GEOMETRICAL ARRAY USED FOR THERMAL ANALYSIS

TABLE B-17

THERMAL PROPERTIES OF GEOLOGIC MEDIA

	Density g/cm^3	Specific Heat $\frac{\text{Cal}}{\text{g-}^{\circ}\text{C}}$	Thermal Conductivity $\frac{\text{Cal}}{\text{cm-hr-}^{\circ}\text{C}}$	Volumetric Heat Capacity $\frac{\text{Cal}}{\text{cm}^3\text{-}^{\circ}\text{C}}$
Salt	2.2	0.22	18-52*	0.48
Granite	2.7	0.21 - 0.26*	22-31*	0.58-0.71*
Shale	2.4	0.20	14.	0.48
Surface Deposit	2.4	0.20	23.7	0.48

---

* Temperature-dependent

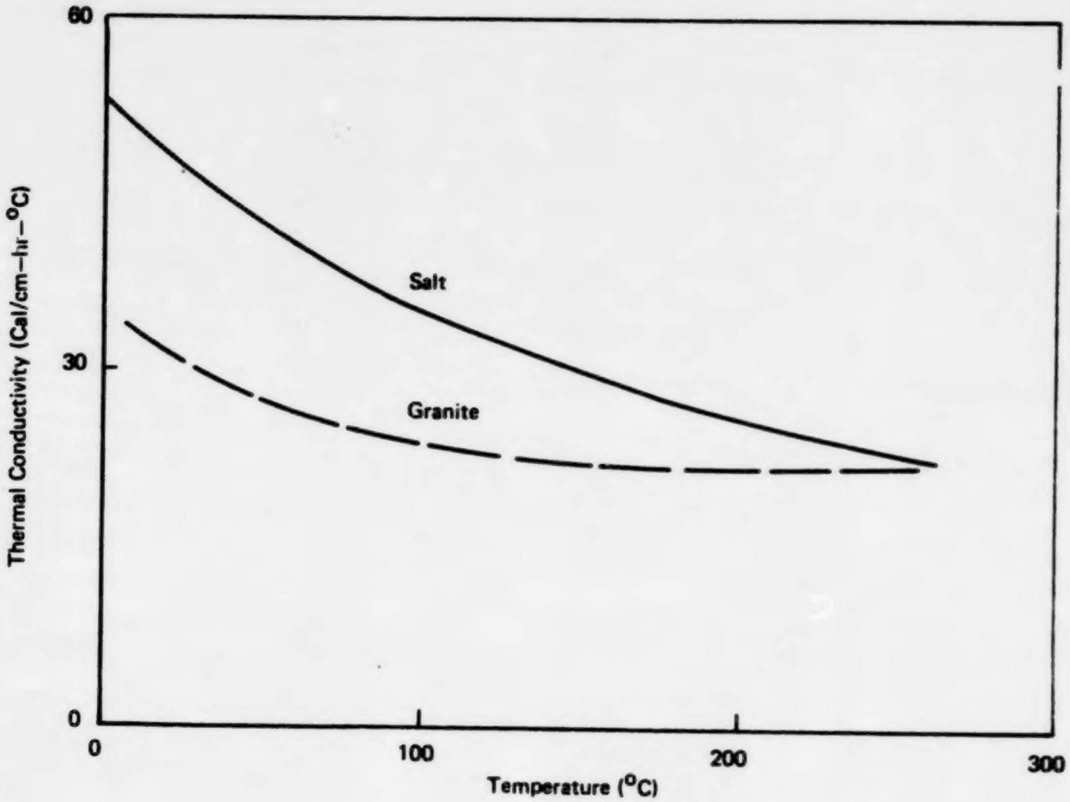


FIGURE B–20 ASSUMED VALUES FOR THERMAL CONDUCTIVITY VS. TEMPERATURE

TABLE B-18

GEOLOGIC FORMATIONS AT DEPTHS IN MODEL

Vertical Positions		I Bedded Salt	II Granite	III Salt Dome
Meters	**Feet**			
0- 150	0- 500	"Surface"	"Surface"	"Surface"
150- 410	500-1350	Shale	Granite	Salt
410- 500	1350-1650	Salt	Granite	Salt
500-1520	1650-5000	Shale	Granite	Salt
1520-2430	5000-8000	Shale	Granite	Granite

Note: In all cases, heat source at 460 m (1500 ft)

Parameters in this study were the burial time, the type of waste (throwaway vs recycled mixed oxide) and the planar heat generation rate at burial time. Cases that were analyzed and their parameters are summarized in Table B-19. The case numbers, such as I-A, III-B, or II-C, are keyed to the indicated geologic formation (Table B-18) and the corresponding heat source (Table B-19).

The results of Cases I-B, II-B, and III-B are given in Figures B-21, B-22, and B-23, showing the temperature rise vs vertical position at 10, 100, 1000, and 10,000 years after burial at a power density of 150 kW/acre in all three geologic formations. The temperature profiles are very similar for all three cases. The highest temperatures are reached between 100 and 300 years, as shown in Figure B-24. The highest temperature was predicted to occur in bedded salt (270°C). The maximum temperature in the granite and in the salt dome was predicted to be about 10°C lower. The slightly lower peak temperature in the granite vs the bedded salt is due to the larger volumetric heat capacity in the granite, which more than offsets the smaller thermal conductivity of granite. The temperature profile above and below the waste containers stays symmetric until the ground surface temperature increases slightly and heat starts flowing from the surface to the atmosphere. A heat transfer coefficient of 0.4 cal/(hr - cm^2 - °C) or 0.8 Btu/(hr - ft^2 - °F) on the surface was assumed for the analysis. The surface temperature increased by less than 0.2°C (0.3°F), and the maximum was reached between 1000 and 3000 years after burial.

The effect of burial time on maximum salt mine temperature is given in Figure B-25, i.e., Cases I-B, I-C, and I-D. It shows the maximum temperature vs time in a 90-m (300-ft) salt layer after burial of 10-, 20-, and 50-year-old waste (recycled mixed oxide) with a power density of 150 kW/acre at the ten-year point. The maximum predicted temperatures are about 250°, 190°, and 120°C for the three cases, reached in 200-300 years.

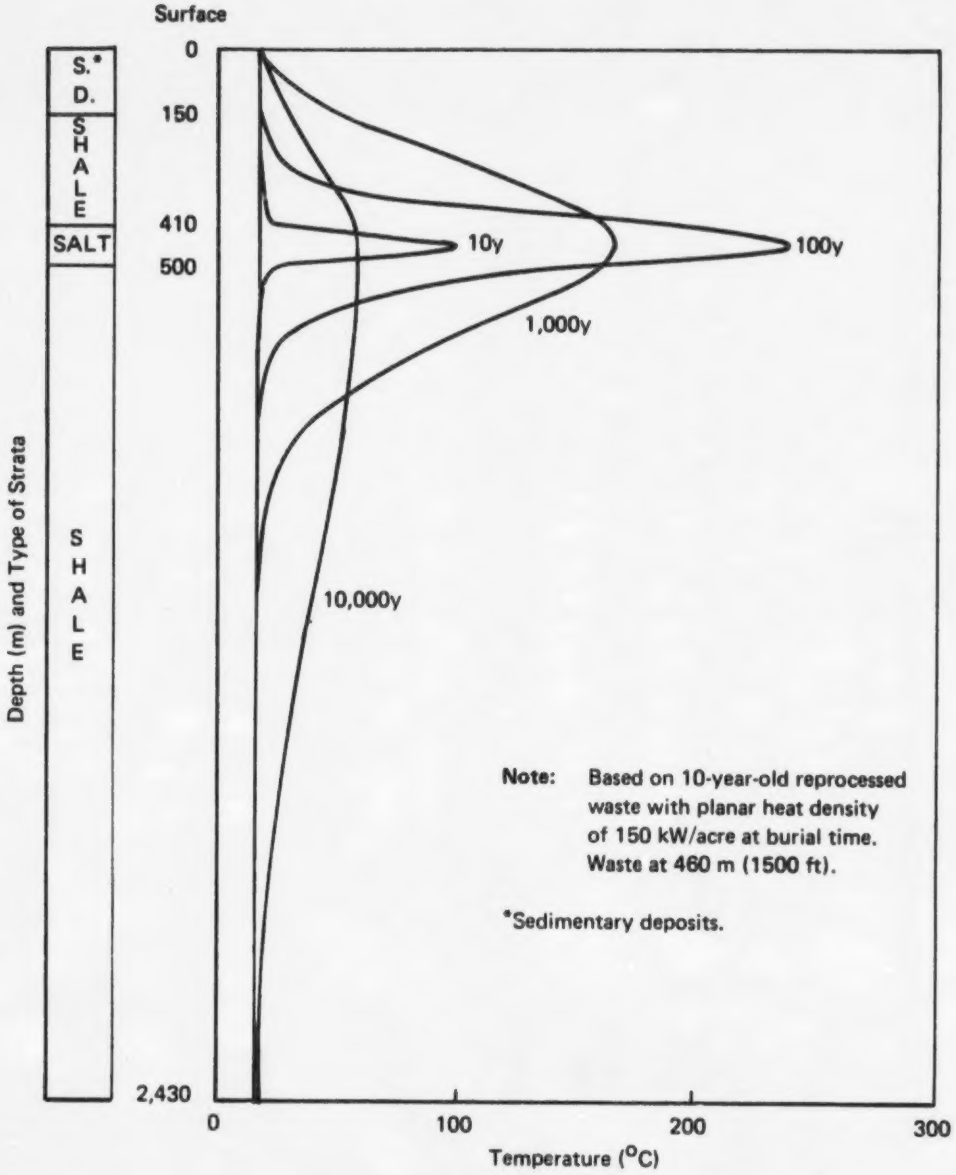
The effect of burial time in granite is shown in Figure B-26 for Cases II-B and II-C. The maximum mine temperature is about 60°C lower for burial of 20-year-old waste compared with burial of 10-year-old waste. The same figure also shows the effect of lower initial power density on temperature rise. Cases II-E and II-F are similar to Cases II-B and II-C, but power density has been reduced for these two cases from 150 kW/acre to 60 kW/acre at the ten-year point. For the lower power density cases, the maximum temperature is reached at about the same time as for the high power density cases. The temperature rise is approximately proportional to the initial power density. For Case II-B, the temperature rise vs vertical position at various times is shown in Figure B-22, while for Case II-E (60 kw/acre) it is shown in Figure B-27.

The last sensitivity run shows the effect of the type of waste on the temperature rise in a salt deposit, with an initial power density of 150 kW/acre at the 10-year point. For throwaway-cycle waste, the predicted maximum temperature is about 60°C higher than for the recycled

TABLE B-19

PARAMETERS AFFECTING THE TIME-DEPENDENT HEAT GENERATION RATE

Case	A	B	C	D	E	F
Waste	Throwaway Cycle	Recycled Mixed Oxide	Recycled Mixed Oxide	Recycled Mixed Oxide	Recycled Mixed Oxide	Recycled Mixed Oxide
Time of Burial, Years after Discharge	10 years	10 years	20 years	50 years	10 years	20 years
Heat Generation Rate at 10 Year Point	$\frac{150 \text{ kW}}{\text{acre}}$	$\frac{150\text{kW}}{\text{acre}}$	$\frac{150 \text{ kW}}{\text{acre}}$	$\frac{150 \text{ kW}}{\text{acre}}$	$\frac{60 \text{ kW}}{\text{acre}}$	$\frac{60 \text{ kW}}{\text{acre}}$



Note: Based on 10-year-old reprocessed waste with planar heat density of 150 kW/acre at burial time. Waste at 460 m (1500 ft).

*Sedimentary deposits.

FIGURE B–21 VERTICAL TEMPERATURE DISTRIBUTION IN TYPICAL BEDDED SALT FORMATION

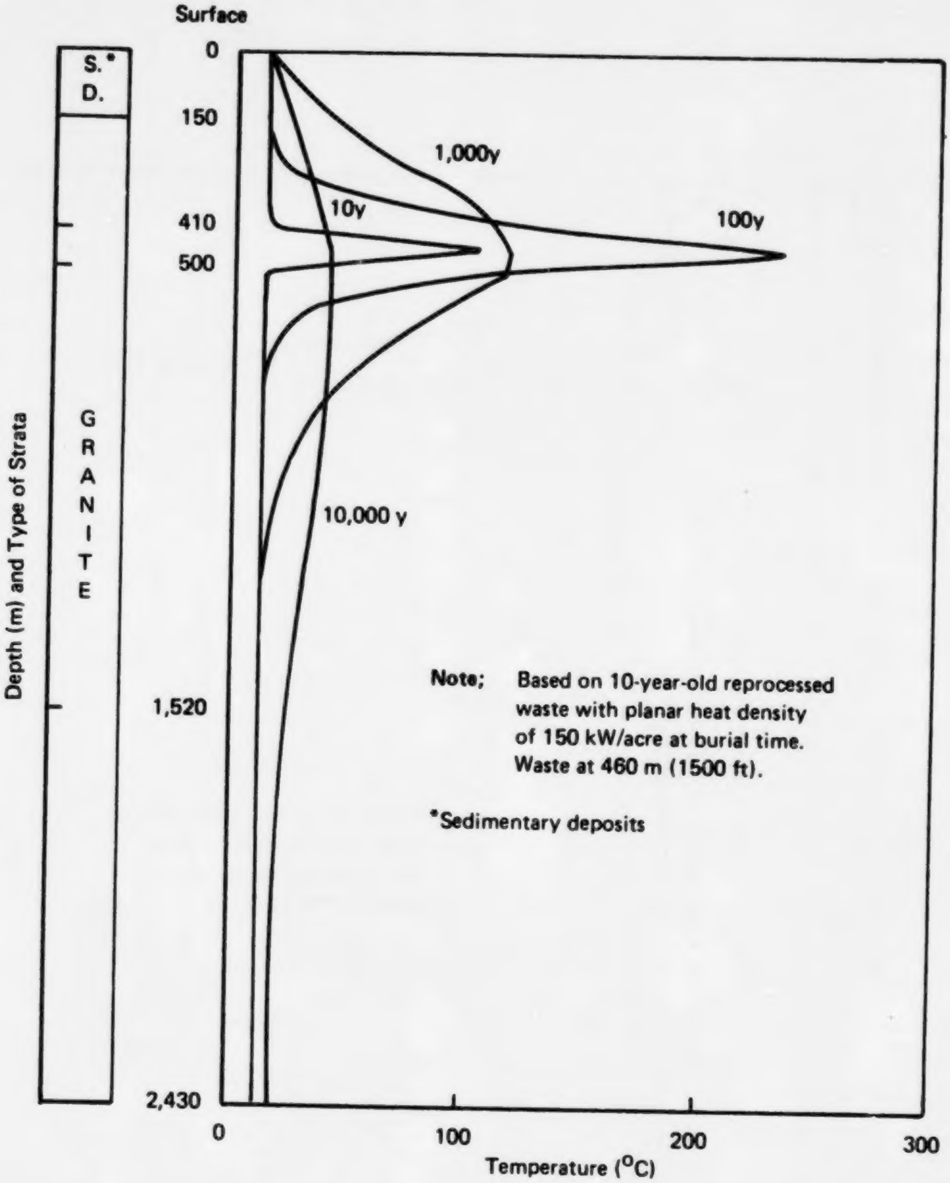
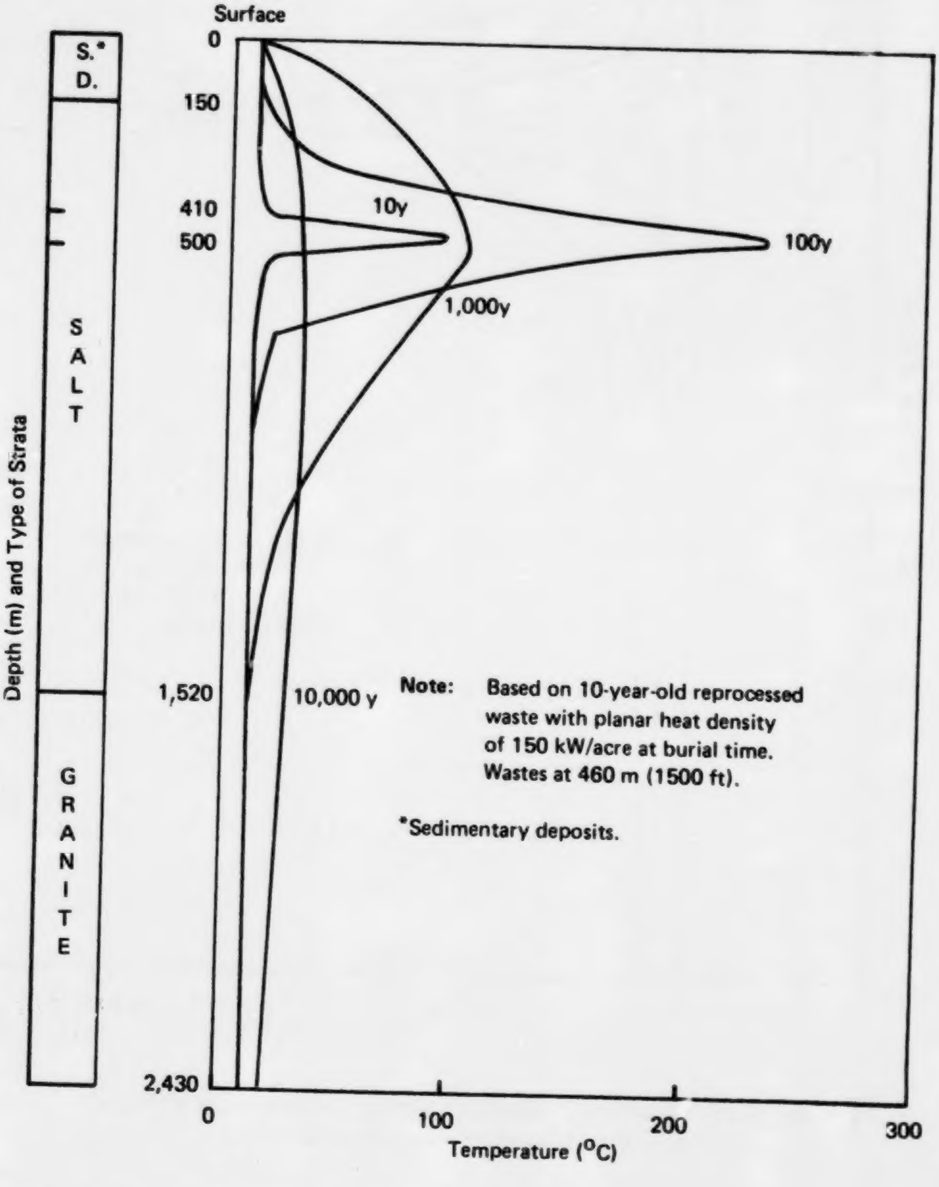


FIGURE B–22 VERTICAL TEMPERATURE DISTRIBUTION IN TYPICAL GRANITE FORMATION (150 kW/ACRE)



Note: Based on 10-year-old reprocessed waste with planar heat density of 150 kW/acre at burial time. Wastes at 460 m (1500 ft).

*Sedimentary deposits.

FIGURE B–23 VERTICAL TEMPERATURE DISTRIBUTION IN TYPICAL SALT DOME

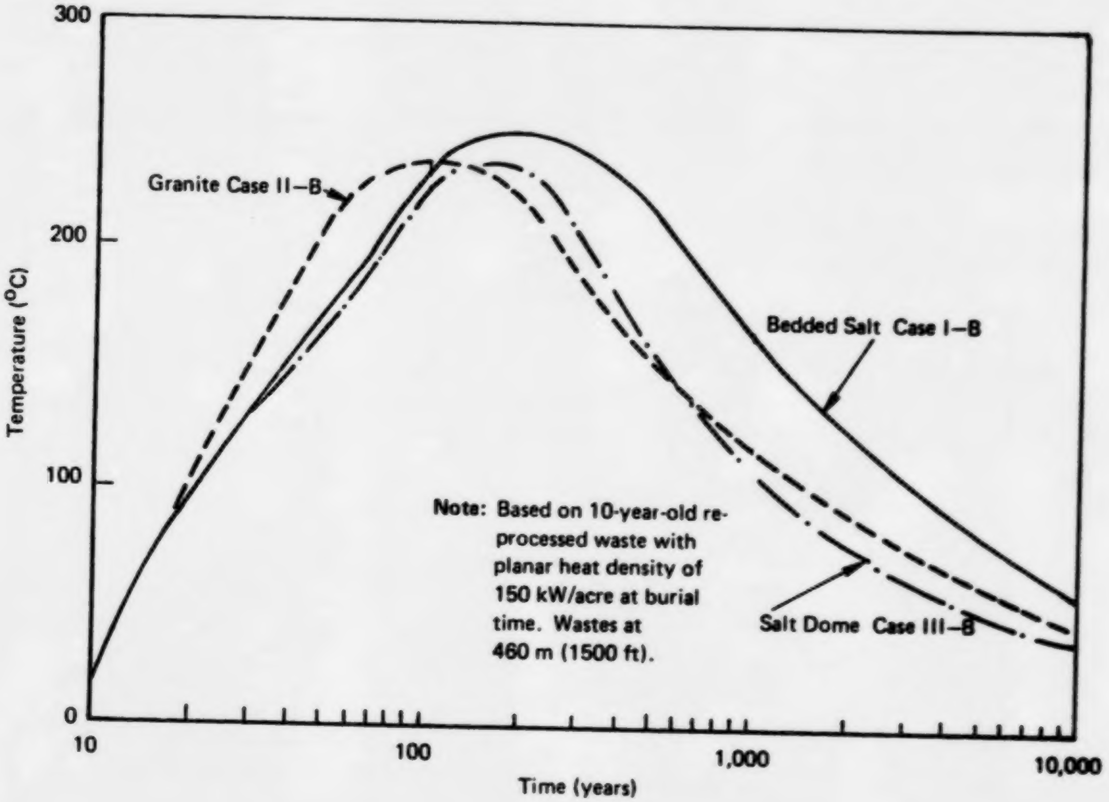


FIGURE B–24 MAXIMUM MINE TEMPERATURE VS. TIME IN THREE GEOLOGIC FORMATIONS (REPROCESSED WASTE)

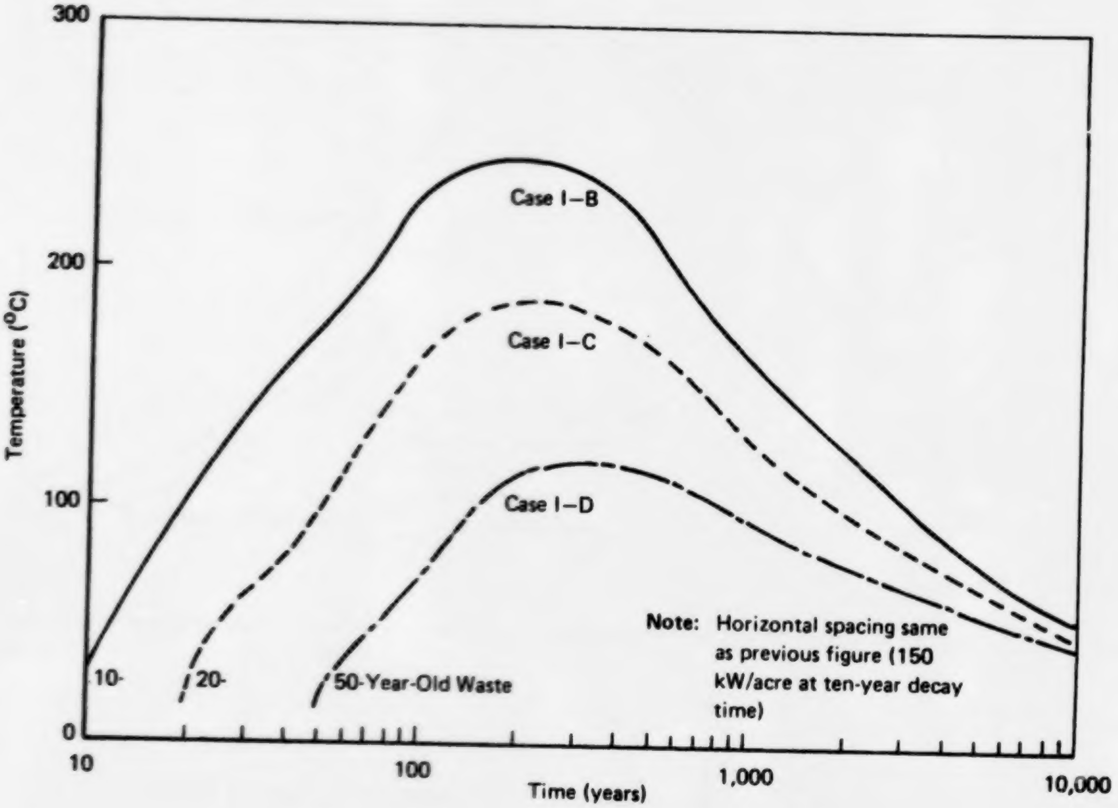


FIGURE B—25 MAXIMUM MINE TEMPERATURE VS. TIME FOR 10-, 20-, AND 50-YEAR-OLD WASTE IN BEDDED SALT

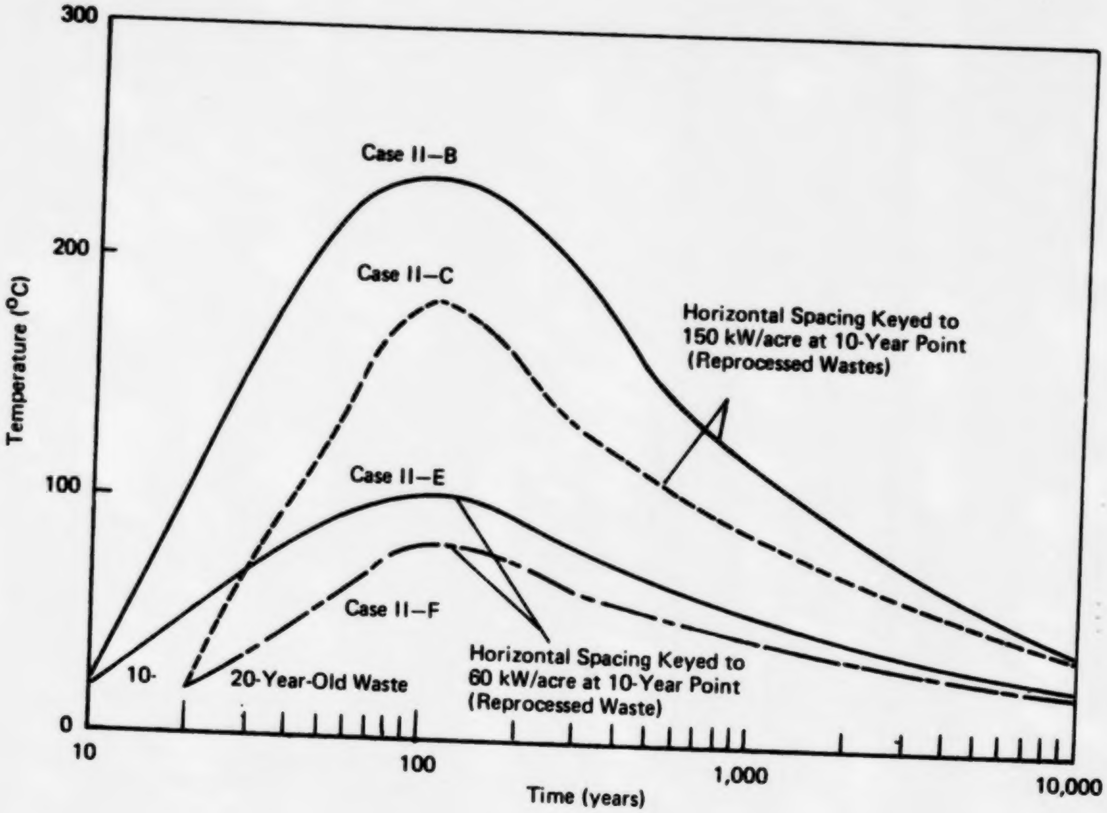
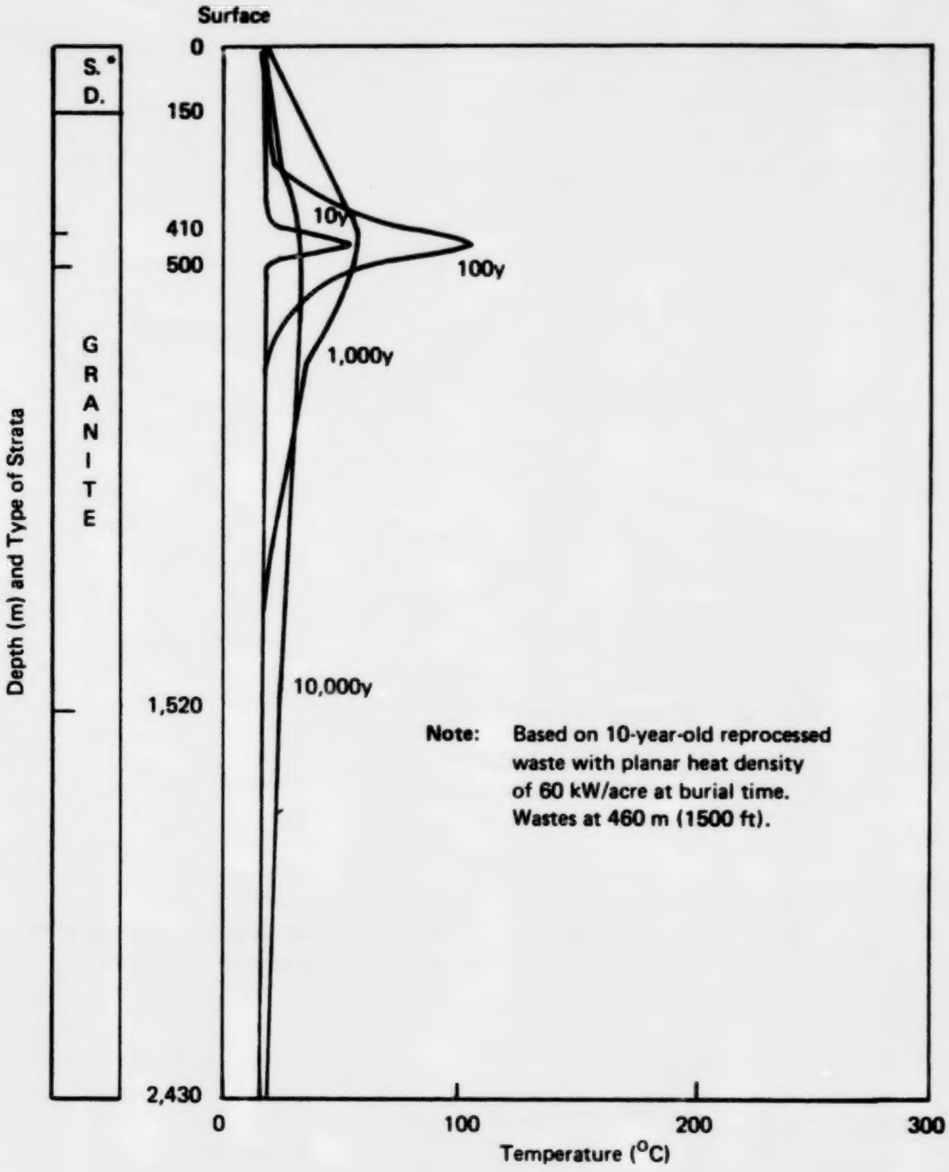


FIGURE B–26 MAXIMUM MINE TEMPERATURE VS. TIME FOR 10- AND 20-YEAR-OLD WASTE IN GRANITE



Note: Based on 10-year-old reprocessed waste with planar heat density of 60 kW/acre at burial time. Wastes at 460 m (1500 ft).

FIGURE B–27 VERTICAL TEMPERATURE DISTRIBUTION IN TYPICAL GRANITE FORMATION (60 kW/ACRE) mixed oxide waste. As shown in Figure B-28, the maximum value is reached at about 170 years for the recycled mixed-oxide waste vs 330 years for the throwaway waste. The higher maximum temperature is due to the fact that the decay heat is sustained for a longer time for the throwaway waste than for the recycled mixed oxide waste.

B-3.5.3.3.2 Results of 3-D Analysis

The objective of the 3-D analysis was to analyze the local temperature distribution and gradients in the salt or granite around the waste containers as a function of time. Because the steepest temperature gradient will occur shortly after burial, since heat flux is highest at that time, and because of the high cost of long-time 3-D calculations, the cases were terminated at the two-year point after burial. Three cases with varying power densities and host materials were analyzed. Heat-generation rates of 3.95 kW/canister and 1.58 kW/canister were used, corresponding to the power densities of 150 kW/acre and 60 kW/acre, the values used in the one-dimensional analysis. The given heat production rates correspond to ten-year-old waste of recycled mixed oxide at the time of burial. The repository material was either salt or granite. The canister was assumed to be buried in the rectangular array shown in Figure B-19. The vertical limit for this 3-D analysis was a 460-m (150-ft) layer above and below the waste container, sufficiently large to cause an insignificant error in the temperature calculation during the time period covered.

The results of the first case, salt with an assumed power density 3.95 kW/canister at burial time, are shown in Figures B-29 to B-31. Figures B-29 and B-30 show the temperature gradient in the salt along the three axes after burial. After two years, the temperature gradient close to the canister is very similar in the X and Y direction and about a factor of two smaller in the Z (vertical) direction. The hot spot after one year is 110°C. Figure B-31 shows the isotherms in the salt at canister mid-plane (Z=0). The isotherms are circular close to the canister, as expected. Farther away, however, the isotherms are parallel to the Y axis.

Figures B-32, B-33, and B-34 show the results of the analysis representing the waste canister buried in granite. The heat production rate was identical to the first case. Less than 3 m (10 ft) from the canister, granite temperatures are significantly higher, and the gradient is about a factor of three larger than in salt. The gradients in the X and in the Y directions are almost identical and a factor of two smaller than in the Z direction. The highest granite temperature at the canister surface is 169°C.

The much higher close-in temperatures for granite than for salt result from substantially lower conductivity (see Table B-17). Farther away, where heat flux is lower and heat storage is more important, the better heat storage capacity of granite (see Table B-17) compensates for

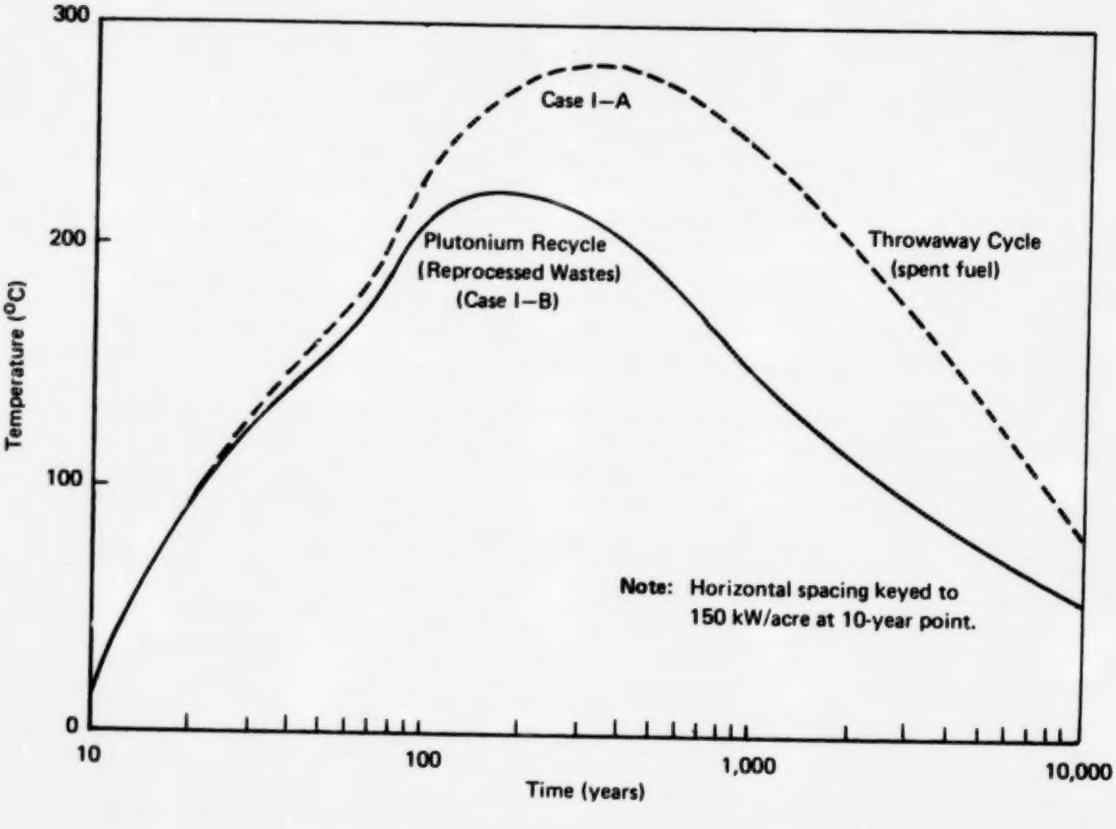


FIGURE B–28 MAXIMUM MINE TEMPERATURE VS. TIME FOR 10-YEAR-OLD WASTE IN BEDDED SALT FORMATION

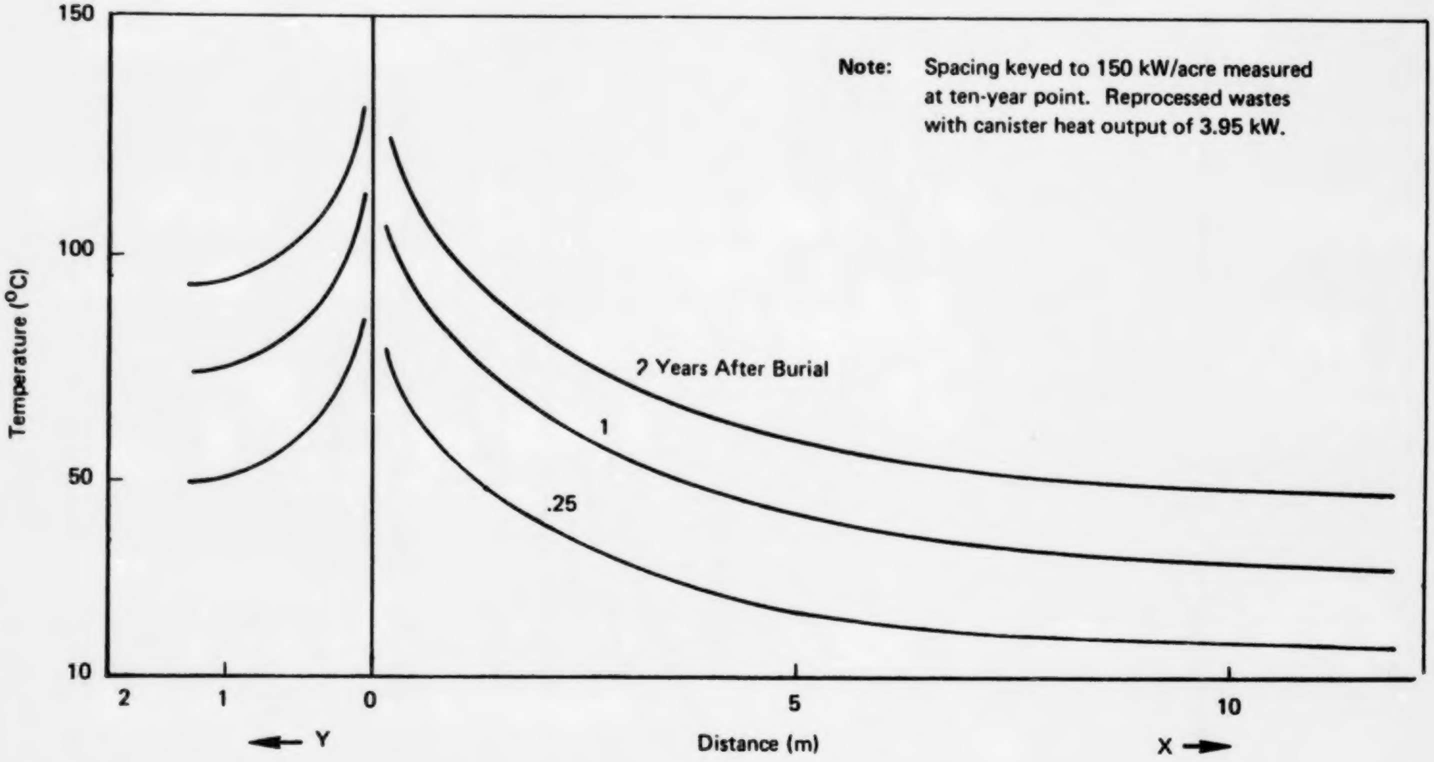


FIGURE B–29 HORIZONTAL TEMPERATURE DISTRIBUTION IN SALT FOR VARIOUS TIMES AFTER BURIAL (3-D CALCULATION)

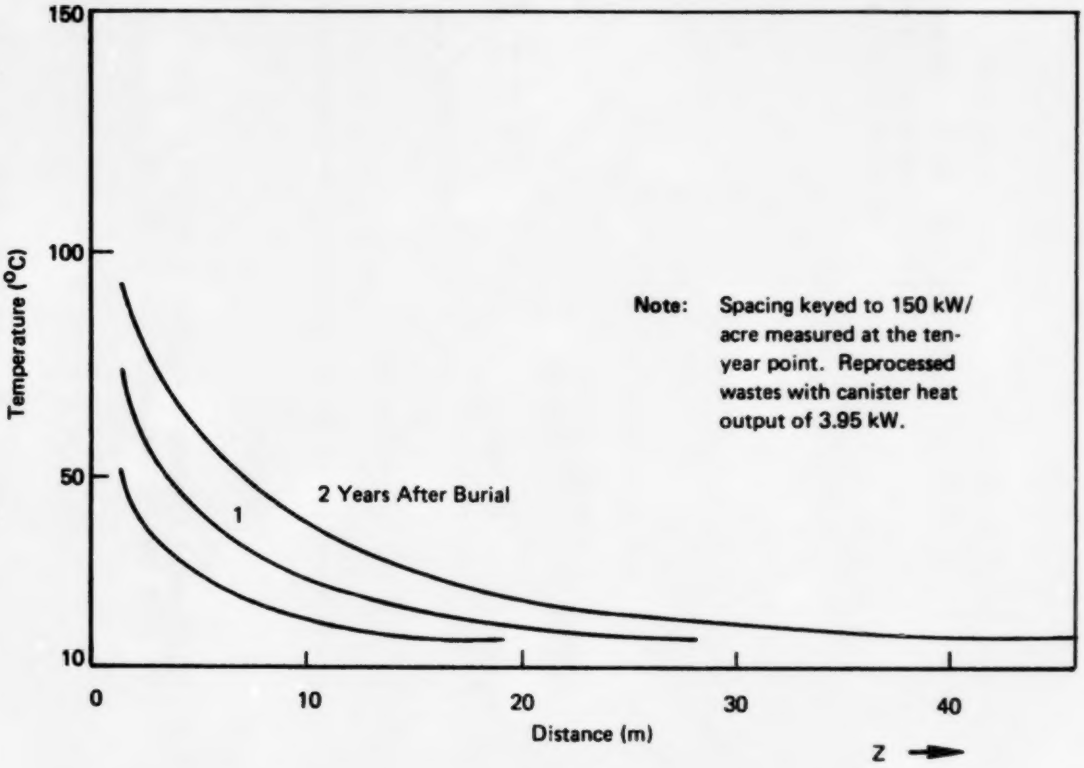


FIGURE B–30 VERTICAL TEMPERATURE DISTRIBUTION IN SALT FOR VARIOUS TIMES AFTER BURIAL (3–D CALCULATION)

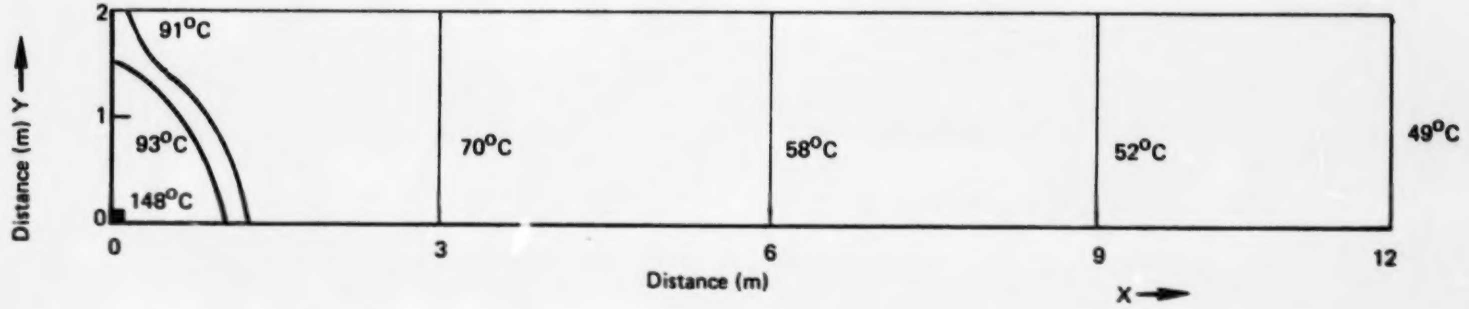
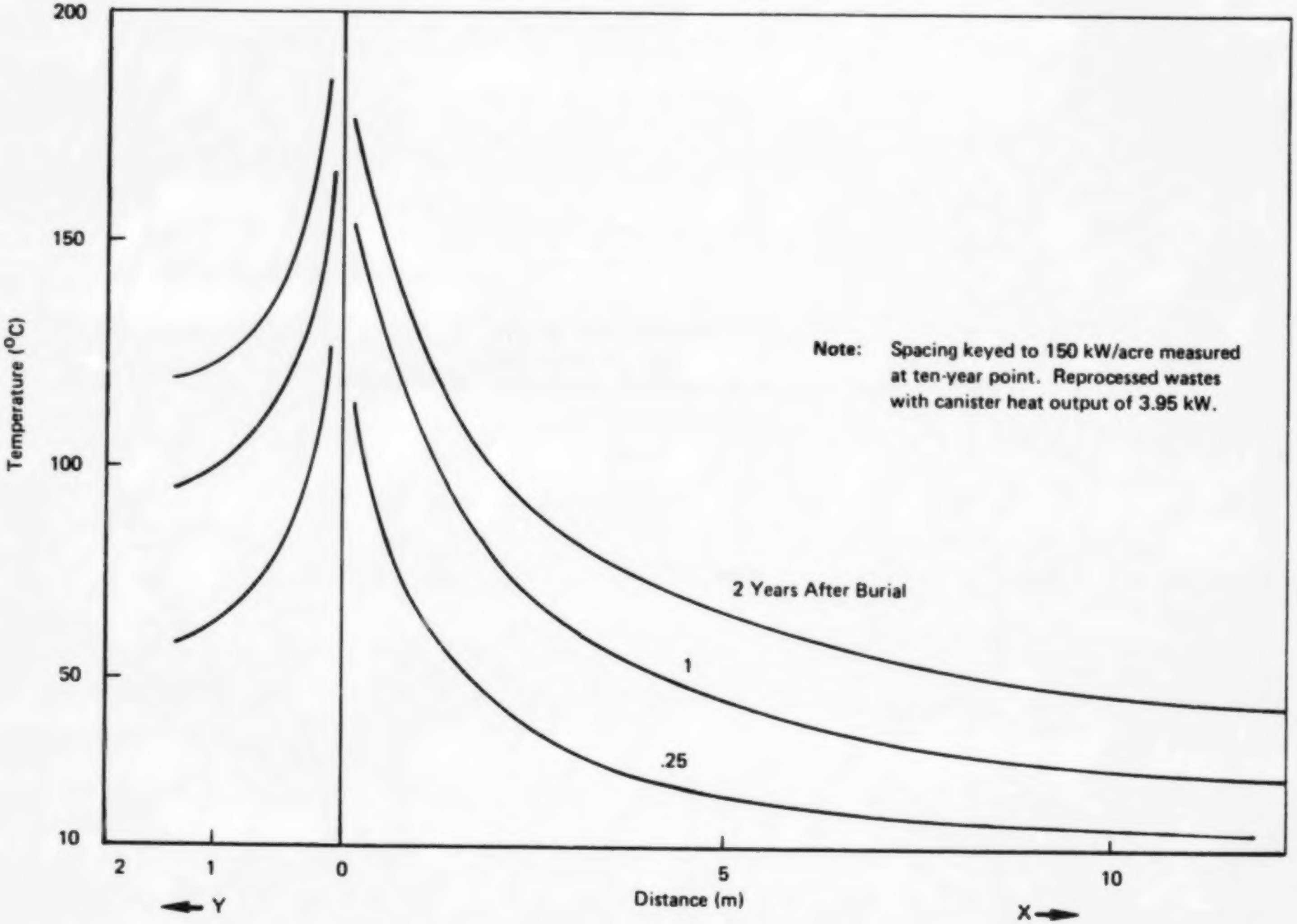


FIGURE B–31 ISOTHERMS IN SALT AROUND THE WASTE CANISTER MIDPLANE (Z = 0) AT THE 2-YEAR POINT AFTER BURIAL (Q=3.95 kW/CANISTER)



Note: Spacing keyed to 150 kW/acre measured at ten-year point. Reprocessed wastes with canister heat output of 3.95 kW.

FIGURE B–32 HORIZONTAL TEMPERATURE DISTRIBUTION IN GRANITE FOR VARIOUS TIMES AFTER BURIAL (3-D CALCULATIONS) (150 kW/ACRE)

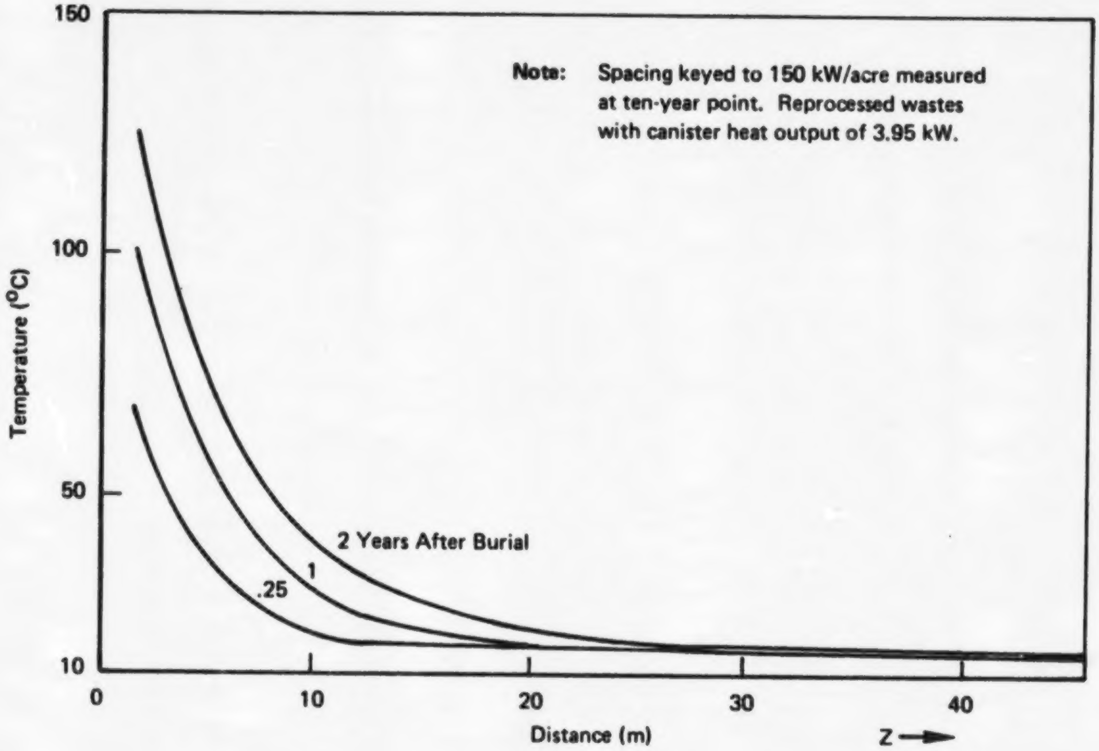


FIGURE B–33 VERTICAL TEMPERATURE DISTRIBUTION IN GRANITE FOR VARIOUS TIMES AFTER BURIAL (3-D CALCULATION) (150 kW/ACRE)

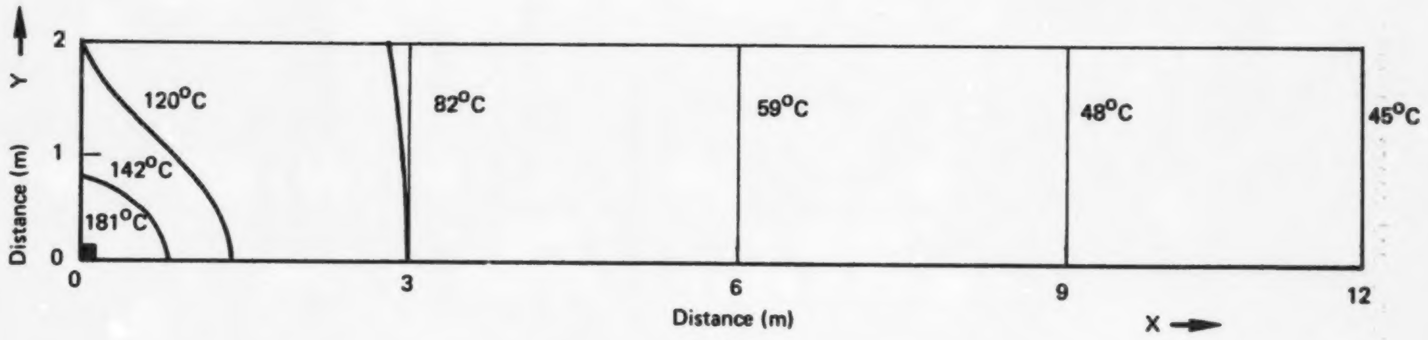


FIGURE B–34 ISOTHERMS IN GRANITE AROUND THE WASTE CANISTER AT CANISTER MIDPLANE (Z=0) AT THE 2-YEAR POINT AFTER BURIAL (Q = 3.95 kW/CANISTER) the lower conductivity, and there is very little difference in the temperature profiles (1-D case).

The effect of a lower heat generation rate in granite (1.58 kW/canister vs 3.95 kW/canister) is given in Figures B-35, B-36, and B-37. The maximum gradient in all three main directions is about a factor of three smaller than for the higher-power case.

No general criteria have been established or suggested for a granite repository. The repository temperature distributions can be tailored to any criteria by varying canister heat output and/or planar heat densities.

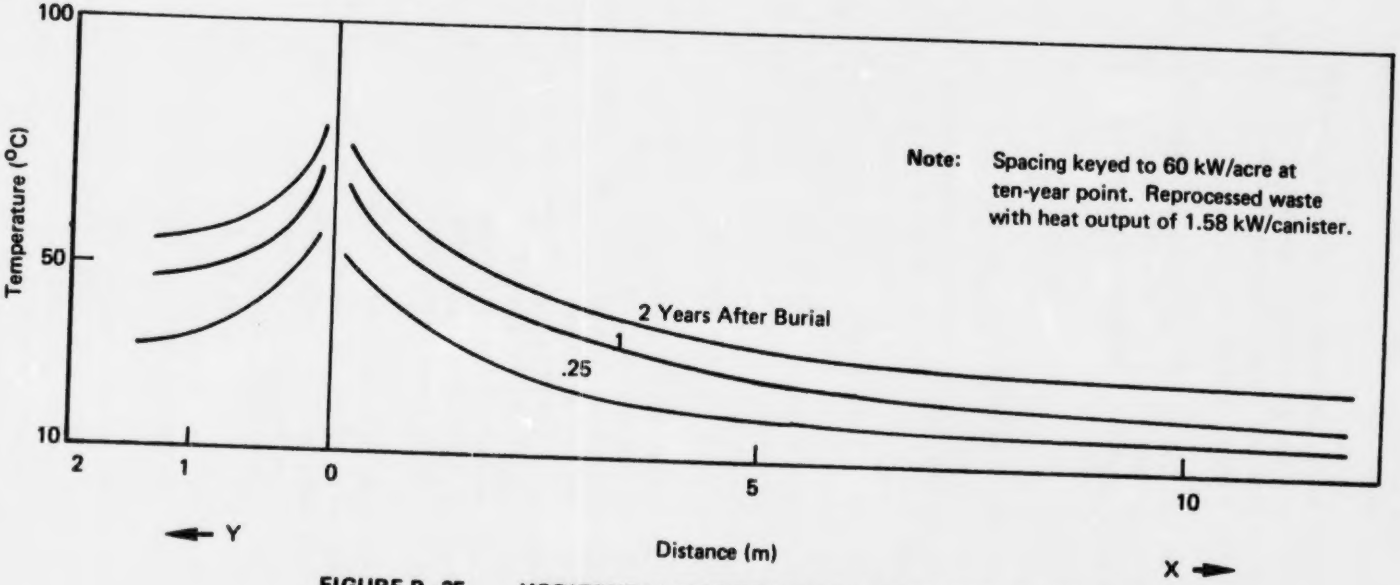
B-3.5.3.3.3 Results of 2-D Analyses

Planar heat densities may be limited by temperature constraints or by thermal expansion and its resultant thermal stresses. Therefore, 2-D calculations have been used to estimate the amount of uplift that might occur in the absence of any constraints on thermal expansion. The 2-D calculations are also necessary in order to examine "edge-effects" at the extremities of the disposal area. (See Figures B-38 and B-39). Based on calculations for a 460-m (1500-ft) repository, maximum stored-heat conditions are reached in 500-1000 years. Maximum uplift is assumed to occur at the time of maximum stored energy.

A summary of the key results of these uplift calculations is presented in Table B-16. For the purpose of this study, it was assumed that all the volumetric expansion, taken as three times the linear expansion, acts in the vertical direction. This is perhaps a reasonable assumption for the relatively plastic salt dome, but for the bedded salt and granite repositories, where little of the expansion is in salt, this assumption probably overstates the amount of uplift. Another element of conservatism is introduced for the salt repositories by virtue of the fact that void collapse (subsidence) has been ignored.

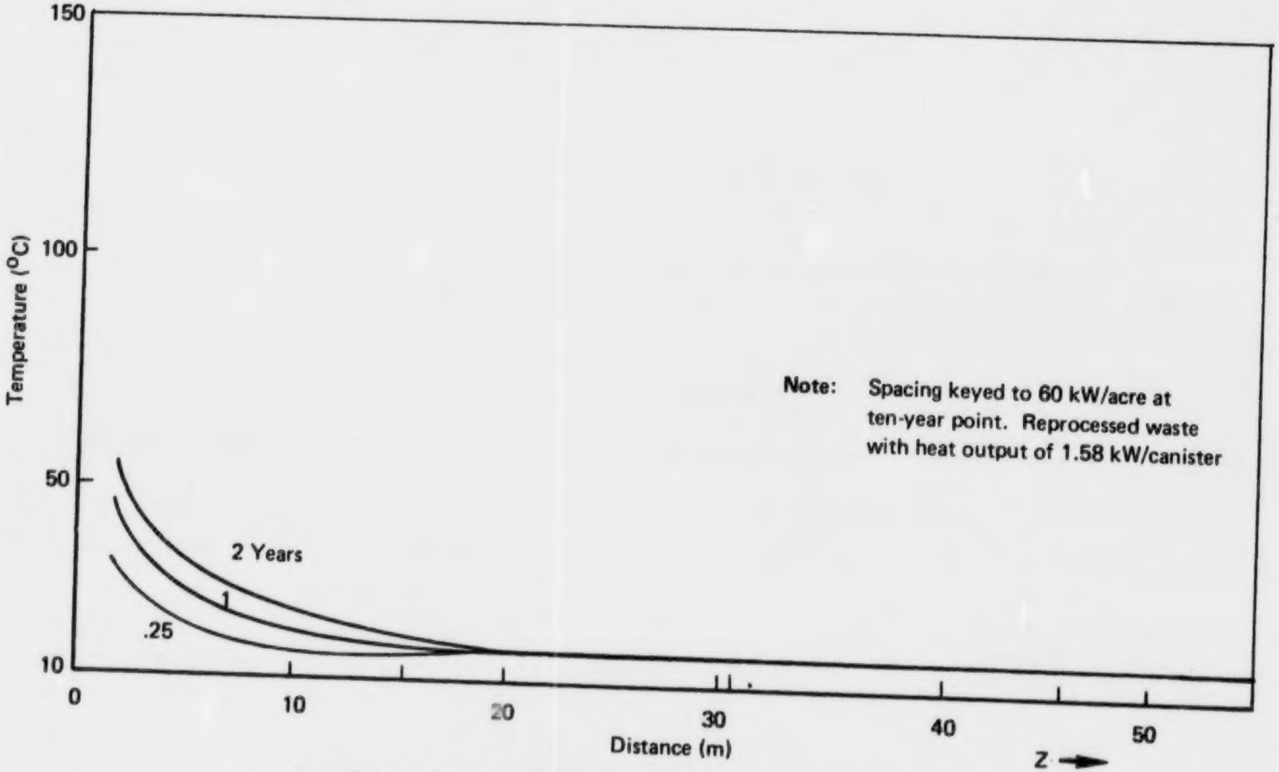
The results of the 2-D calculations, based on the same volumetric expansion coefficients, are shown in Figure B-40. The calculated uplift is quite uniform across the central portion of the storage area, but drops off rapidly at the boundary. Even with this rapid drop-off, however, the calculated maximum ground slope near the boundary is rather small, only 1.5°, 0.5°, and 0.2° for the salt dome, bedded salt, and granite repositories, respectively. The calculated uplift decreases to zero about 2000 feet outside the repository boundary.

Recognizing that these uplift values are conservative, the edge effects do not appear to represent a major problem. If necessary, planar heat densities can be lowered at the edges of the repository to smooth out edge effects. Nevertheless, there are still uncertainties, especially for granite; these relate to the effect of uplift and/or thermal stresses on the integrity of the rock above the repository. Specifically, it



Note: Spacing keyed to 60 kW/acre at ten-year point. Reprocessed waste with heat output of 1.58 kW/canister.

FIGURE B–35 HORIZONTAL TEMPERATURE DISTRIBUTION IN GRANITE FOR VARIOUS TIMES AFTER BURIAL (3-D CALCULATION) (60 kW/ACRE)



Note: Spacing keyed to 60 kW/acre at ten-year point. Reprocessed waste with heat output of 1.58 kW/canister

FIGURE B–36 VERTICAL TEMPERATURE DISTRIBUTION IN GRANITE FOR VARIOUS TIMES AFTER BURIAL (3-D CALCULATION) (60 kW/ACRE)

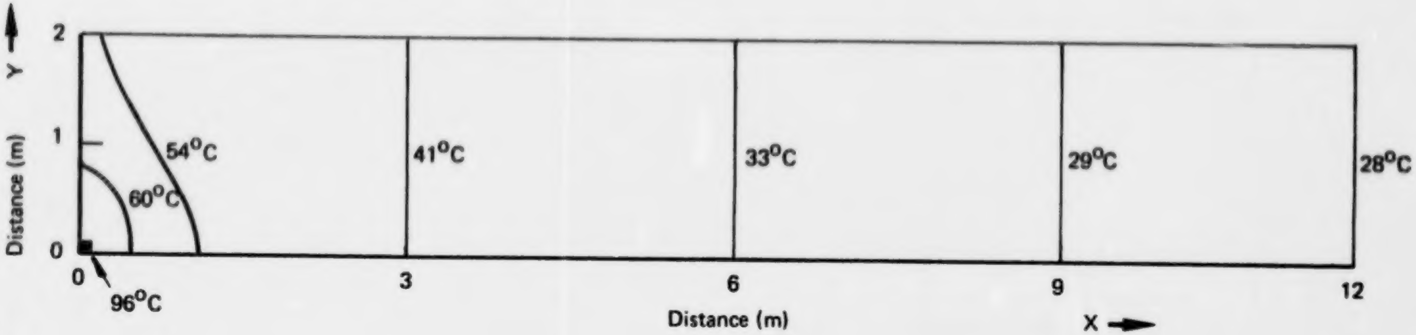


FIGURE B–37 ISOTHERMS IN GRANITE AROUND WASTE CANISTER AT CANISTER MIDPLANE (Z = 0) AT THE 2-YEAR POINT AFTER BURIAL (Q = 1.58 kW/CANISTER)

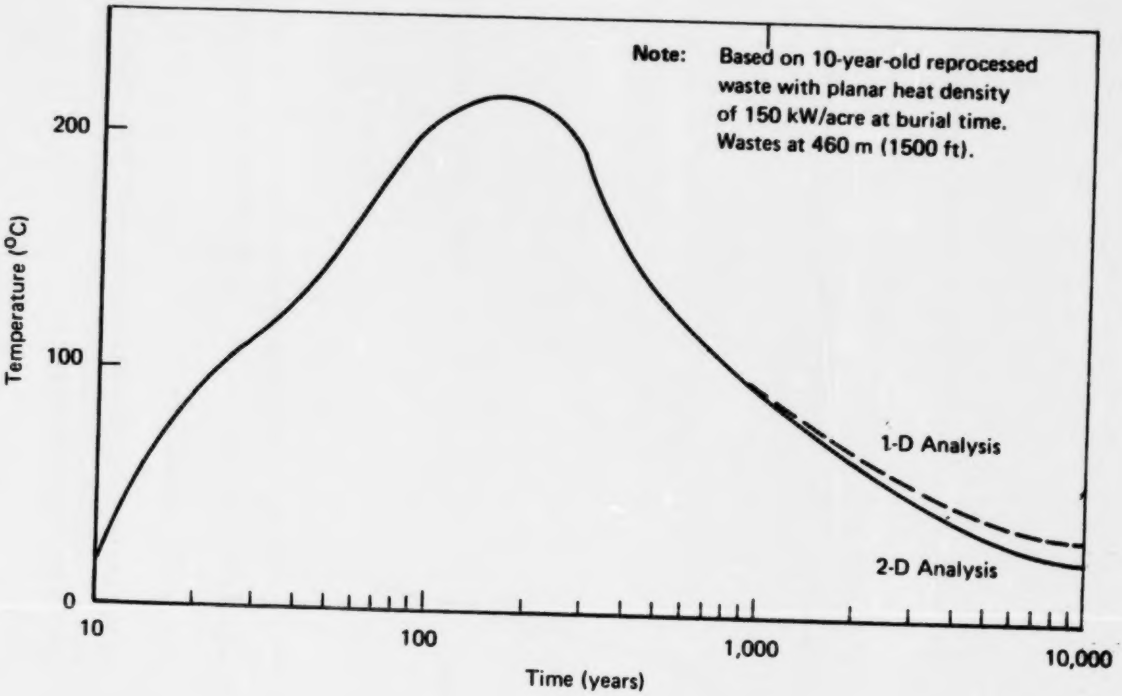
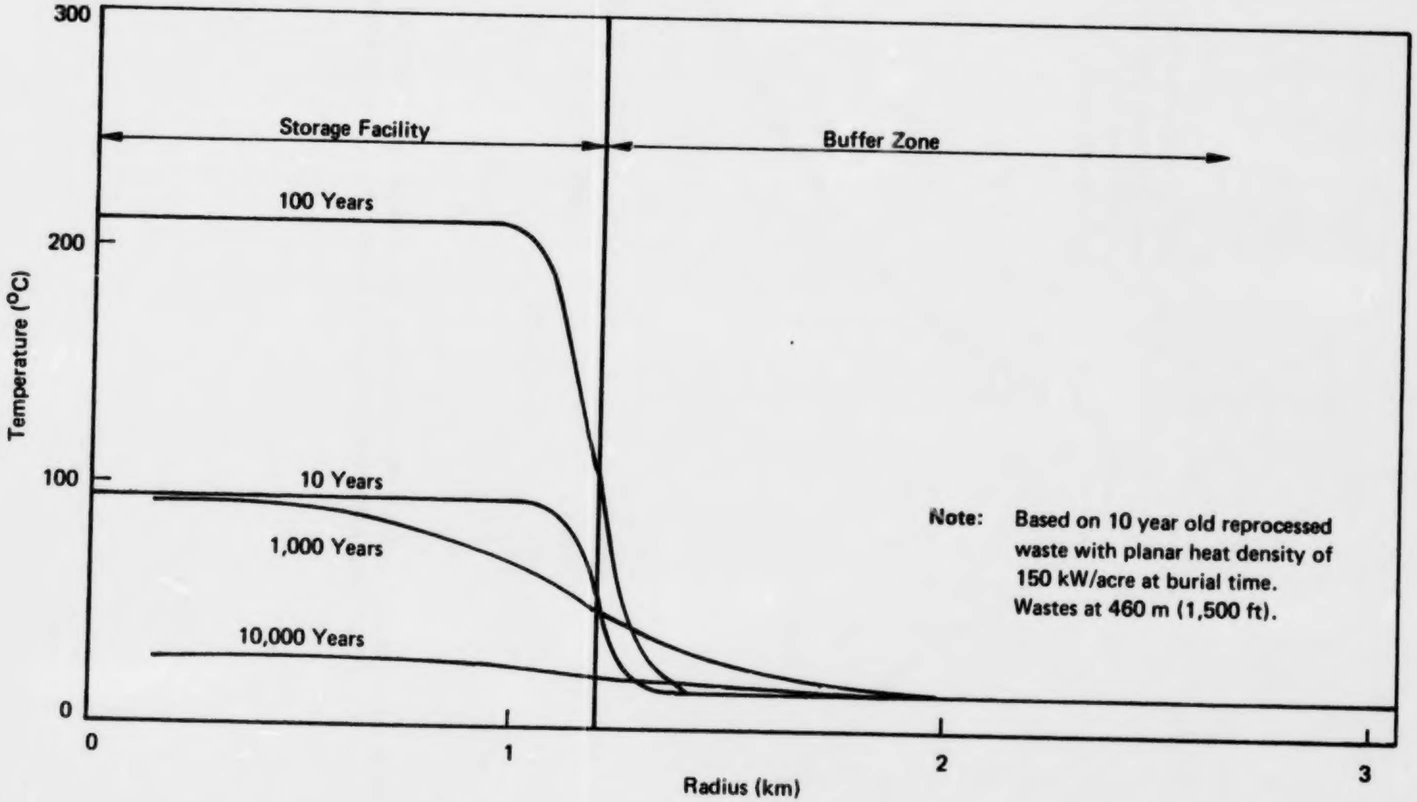


FIGURE B–38 COMPARISON OF PREDICTED MAXIMUM MINE TEMPERATURE VS. TIME FOR A ONE- AND TWO-DIMENSIONAL ANALYSIS IN SALT DOME



Note: Based on 10 year old reprocessed waste with planar heat density of 150 kW/acre at burial time. Wastes at 460 m (1,500 ft).

FIGURE B–39 TEMPERATURE RISE VS. RADIUS AT CANISTER MID-PLANE (Z = 0) FOR A 1100-ACRE STORAGE FACILITY IN A SALT DOME (RESULT OF 2-D (RESULT OF 2-D ANALYSIS)

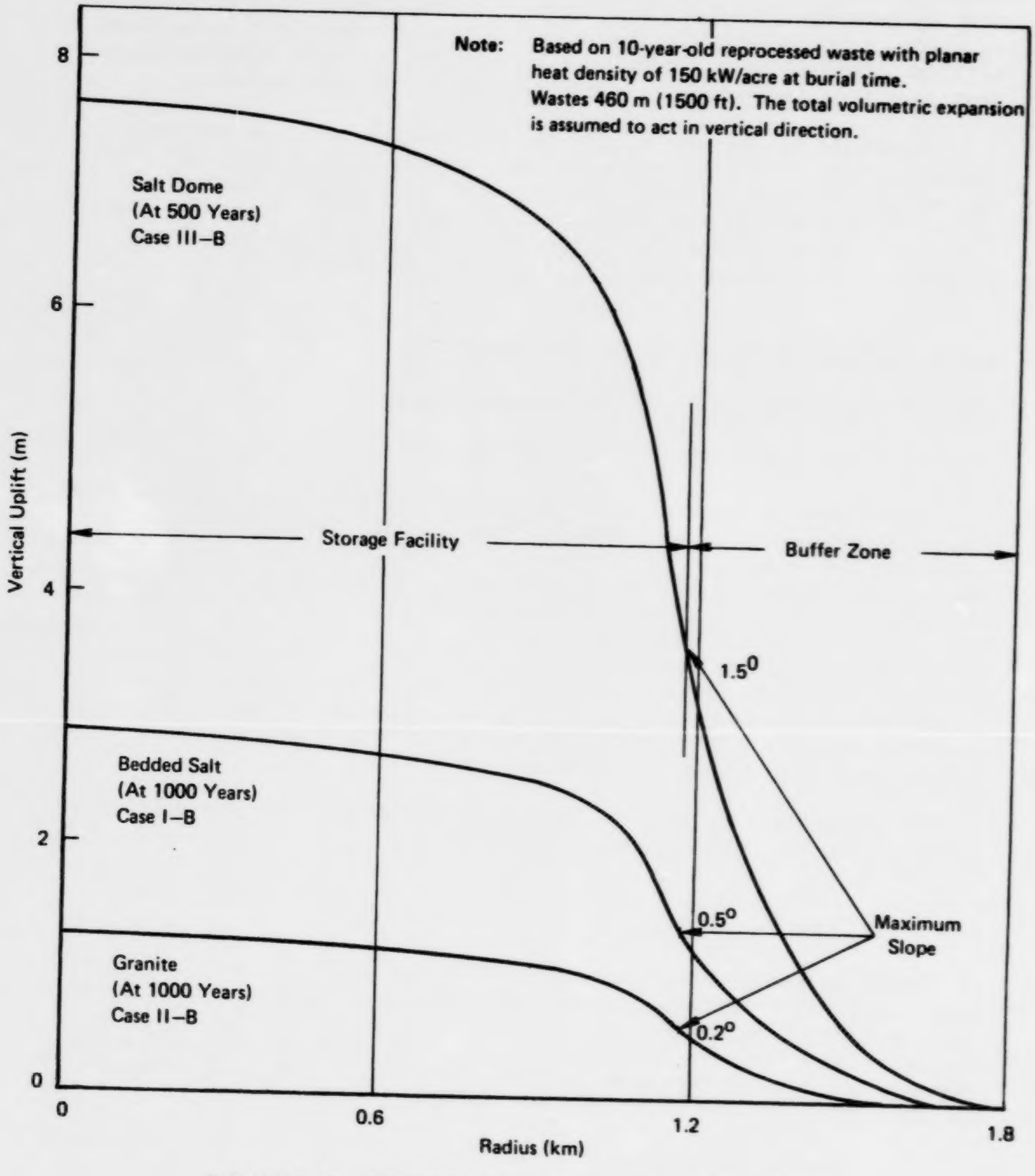


FIGURE B–40 MAXIMUM VERTICAL UPLIFT VS. RADIUS FOR A 1100-ACRE STORAGE FACILITY IN THREE GEOLOGIC FORMATIONS (RESULTS OF 2-D ANALYSIS) would be undesirable if thermal stresses caused connected faulting to develop between the repository and near-surface rock; further work is needed on rock mechanics to determine whether this is a significant problem. This potential problem can be accommodated by limiting planar heat densities sufficiently, and by smoothing out edge effects.

It should be emphasized that the preceding discussion addressed thermal uplift problems for HLW burial. For spent fuel disposal, the maximum stored energy at 500 years is about twice that of high-level waste. This difference is best noted in Figure B-28. Thus, for the same limits on total uplift, planar heat densities would have to be roughly a factor of two less for spent fuel than for HLW.

B-3.6 EFFECTIVENESS OF ENGINEERING CONTROLS

B-3.6.1 Reference Technologies in Bedded Salt

B-3.6.1.1 Barriers to Transport from Repository

Since even in intact salt there is perhaps 0.1-0.5% water and since the water tends to migrate toward a heat source, the high-level waste canisters are assumed to be exposed to saline waters that can corrode the canister from the time of emplacement. Subsequently, in the case of HLW glass, slow leaching and removal of the glass itself would occur. With simple calcines, dissolution is almost immediate for practical purposes after the canister is destroyed. In spent fuel disposal, after the canister, the fuel cladding, and finally, the uranium oxide of the fuel itself will leach away. In each case, the rate of removal of actinides from the repository may be limited by the low solubility of the actinides in the water flow through the repository.

The immediately following sections discuss canister destruction, leach rates, and actinide solubility limits.

B-3.6.1.2 Waste Canister Integrity

The importance of the integrity of a canister to the transport of high-level wastes from the repository will depend upon the contents of the canister. For example, for a canister containing HLW glass, the canister integrity may be of minor importance because, even with a rapid destruction rate of the canister, the rate of radioactivity release will be limited by the low leach rate of the glass.

Conversely, with a more leachable calcine, canister integrity could be important, especially over a period of several hundred years.

For the glass reference case, the canister material will probably be determined by the requirements of the glass loading and handling operations. As reviewed in Section B-3.2.1 of this report, the "in-can" melting process requires a stainless steel canister or other high-strength alloy material (e.g., Inconel).

For calcine or spent fuel in salt, it is not at all clear that stainless steel canisters would suffice. The integrity of the canister is needed for a long period, but the high temperature and the water present in salt would tend to subject the stainless or high alloy steel to stress chloride cracking. This potential for rapid destruction suggests that, for calcine or spent fuel, some other canister material, e.g, titanium, be utilized. For titanium in the presence of salt and other chloride-containing compounds, even at fairly high temperatures, data indicate a corrosion rate of about 0.025 mm (1 mil) per year (see Appendix B-V). Much better corrosion rates could be possible for specifically-tailored titanium alloys but these may not be cost-effective. It seems clear that, with a relatively modest titanium outer canister (e.g., 13-mm (0.5-in) titanium), the integrity of the canister could be about 500 years. A somewhat thicker canister, perhaps 25 mm (1 in), could be hypothesized to last about 1000 years, even if the "poor environment" were to develop immediately after the high-level waste repository is sealed.

This kind of time frame, 500 to 1000 years, is critical for the easily-leachable calcine; during this period the initially controlling hazard, fission products, decays to a level that is unimportant compared with that of the long-lived transuranic isotopes. The two fission products of major concern are cesium-137 and strontium-90, which have half-lives of 30.2 and 28.9 years, respectively.

B-3.6.1.3 Leachability of Matrix

B-3.6.1.3.1 General Considerations

The leachability of high-level waste glasses will be significantly influenced by several conditions, many of which are unknown and to a degree, unpredictable. Considering that a wide range is possible, the estimate developed should be a high one, where there is little likelihood of a higher removal rate.

Several of the parameters that bear on glass leachability are:

1. The type of glass and its loading of waste solids.
2. The chemical characteristics of the leaching water.
3. The temperature and pressure of the leach system.
4. The physical size characteristics (surface-to-mass ratio) of the glass.
5. The aging characteristics of the glass in the presence of radiation.

The above factors were considered, the literature reviewed, and technical discussions held, in order to understand the current status of technology and to develop best estimates that are related to the above effects. These points will be discussed in more detail below and the reference values for the in-situ glass waste storage system will be suggested.

Besides the HLW glasses, two other alternatives are considered. The first is that of a calcine of the high-level waste (see Section B-3.2.1.1.1), where the leach rates are so high that calcine failure can be considered to take place over a few days' time, once the canister integrity is breached. The other form of high-level waste to be considered is that of spent fuel assemblies (see Section B-3.2.5), which appear capable of behavior anywhere between that of a glass or a calcine. The bases for the development of the reference leach rate limits will be reviewed below.

B-3.6.1.3.2 Glass Leach Rates

In choosing an appropriate leach rate for glass, a fundamental question is how well the various laboratory tests used to evaluate this parameter reflect the behavior of such glass in a long-term geologic environment. Furthermore, the procedures used for the tests themselves can, and do, sometimes give substantially different results for a number of reasons. One reason is related to whether the leaching process is one in which water diffuses into the surface of the glass and removes specific elements, or whether layers of the glass itself dissolve and with it the elements contained in those layers, a process sometimes described as a "corrosion effect". In the short-term laboratory tests, the leach phenomenon appears to be a combination of these two mechanisms. Such a combination explains a faster leach rate for a few days of leaching and a much lower leach rate that appears to be approaching an equilibrium removal condition after longer periods, e.g., years. On the other hand, even a few years is a short period of time compared with the geologic time involved in the dissolution of glass after burial.

The data are further clouded by differences in the character of the leach water, the temperature, and the physical (mechanical) flow at the surface of the glass. Except for a long-term field test in Canada(37) very little data exist to evaluate an actual storage situation, and no results have yet been published for hot salt solution.

Laboratory measurements of leach rates are generally made by measuring a particular radioisotope in water after it has been in contact with a glass for a given period of time. On the basis of the original concentration of that particular isotope in the glass, a corresponding average glass leach rate (corrosion) can be calculated. If this removal is a "corrosion" effect, or removal of a layer from the surface of the glass, it would not matter which isotope was used for the measurement, as any isotope would represent the same fraction of glass removed. On the other hand, if the release were a diffusion removal phenomenon, one element might be leached preferentially to another and, if calculated as above, would give a higher glass leach rate. Furthermore, a short leach time could be surface diffusion-controlled while, as explained above, a long leach period could be more corrosion-related. Most tests, however, are usually limited to periods of much less than one year. Several different sources of leach data have been reviewed. Table B-20 summarizes the range of rates found, depending upon several of these conditions.

Both the Mendel data and the Ross data in Table B-20 indicate a leach rate of 10^{-6} g/cm^2-day at the one-day point and about 10^{-7} g/cm^2-day at the one-year point.(38,39) These values are consistent with the 10^{-5} to 10^{-7} range given in an international summary paper(11), all apparently based on room-temperature leach testing utilizing an International Atomic Energy Agency (IAEA) type of experiment. Ross also indicates an early leach rate of approximately 10^{-5} g/cm^2-day but in a different (Soxhlet) type test apparatus. The Soxhlet type of equipment basically distills water, condenses it, and flushes the hot water over the surface of a sample. This kind of test is substantially different from the IAEA procedure in several details:

1. It is at an elevated temperature, e.g., approximately 100°C.
2. The water in contact with the sample has been distilled and condensed so there is no solid content in the water that can inhibit leachability.
3. Because of the flushing, there is more agitation of the liquid at the sample surface, which would accelerate leaching.

The Merritt data in Table B-20 are of interest because they summarize laboratory and field data on the same material.(37) Two laboratory tests were run on the basic glass used in the experiment, a nepheline syenite glass; the second one, Glass 2 in Table B-20, is applicable to the field test.

When this glass was put into the ground in a closely-packed array, in an area where ground water flowed rapidly, the initial leach rate was about 4×10^{-8} g/cm^2-day. It decreased rapidly until at about seven years the value leveled out at 5×10^{-11} g/cm^2-day. The factors that specifically relate to this large reduction cannot be identified. Furthermore, there are apparently no other field tests from which a generalized conclusion could be evolved. These Canadian tests were at conditions substantially different from those expected in a salt repository.

Recent experiments at Pennsylvania State University(1), conducted at extreme conditions (300°C, 300 atmospheres in the presence of water) show that glass may be destroyed in a matter of weeks; the effect of this information in limiting the thermal loading of repositories has not yet been addressed.

TABLE B-20

GLASS LEACH RATE CONSTANTS - Gm Glass/cm^2-day

Source:	Leach Period - Days					
	1	20	115	300	700	2900
Mendel(A)						
Based on Cs and on Sr	10^{-6}		10^{-7}			
Ross(B)						
Based on Soxhlet Test: 10^{-5} (Time not indicated)						
Based on Cs	10^{-6}	$4x10^{-7}$	$2x10^{-7}$	$1.5x10^{-7}$		
Merritt(C)						
First Test (Glass 1)	10^{-5}	10^{-7}		10^{-8}		
Second Test (Glass 2)	10^{-5}	10^{-6}		$7x10^{-8}$		
Field Test (Glass 2)	$4x10^{-8}$				$4x10^{-10}$	$5x10^{-11}$

(A) High-Level Waste Glass - Mendel, E., Nuclear Technology, Vol. 32, January 1977. Devitrification on these same glasses increased leach rate by a factor of 10 - 20.

(B) Properties and Characteristics of High-Level Waste Glass, Ross, W.A., BNWL - SA-6146, Washington, D.C. Soxhlet test is batch "flowing" water at approximately 99°C. Devitrification increased leach by up to six times as measured by Soxhlet test.

(C) High-Level Waste Glass: Field Leach Test, Merritt, W.F., Nuclear Technology, Vol. 32, January 1977.

Thus, the leach data for glasses are conflicting, imprecise, and scanty. The effects of temperature, flow, pH, and salinity have not even begun to be studied in an organized way for the spectrum of candidate glasses. Laboratory tests to date have not been lengthy enough to achieve a measure of the long-term (steady-state) corrosion rate. Theoretical studies(40) indicate that these rates should continue to decrease with time for considerable periods, a conclusion that is not inconsistent with the results of the one long-term field test reported in Table B-20.

It appears unreasonable to do more than assume a conservatively high value for leach rate constant, allowing for the uncertainties in the above parameters. (The assumption has been made that temperature affects the leach rate as it does any diffusion-controlled phenomenon -- i.e., that the rate varies with the square root of the absolute temperature. Under this assumption, even at 200°C the rate would be only 26% faster than at 25°C. The relatively minor effect of temperature seems supported by the Soxhlet data in Table B-20.) A value of 10^{-6} g/cm^2-day has been chosen because it is at least a factor of 7 to 100 times larger than any data reported after one year. By not assuming it to decrease with time as do the laboratory values, a further conservatism is introduced into this value, which should more than compensate for any uncertainty regarding devitrification (see below). At the same time, it must be admitted that the data indicate that it is entirely possible that leach rate constants as low at 10^{-8} g/cm^2-day may be achieved in practice. Accordingly 10^{-6} g/cm^2-day has been adopted as a reasonably conservative long-term value, and 10^{-8} g/cm^2-day as a possibly achievable long-term value.

B-3.6.1.3.2.1 Surface-to-Mass Ratio

The key determinant in establishing a particle size for the high-level waste glass deals with the glass "casting" operation and its subsequent cool-down rate. After the glass is poured into the waste disposal canister, or alternatively, after the glass melting operation takes place in the disposal canister, cool-down can be rapid until the glass reaches a temperature of approximately 500-600°C, at which point the glass is still plastic enough that cracking will not occur. Below this temperature the cool-down rate, in combination with the shrinkage stresses, will determine the amount of cracking that ultimately takes place in the glass.

This effect has been quantitatively evaluated by BNWL, which indicates that the relative surface areas of the as-cast cylinders, in this case 15 cm (6 in) diameter, increased by a factor of 20-25 for rapid water quenching, a factor of 12 for air-cooling, and a factor of about 5 for slow-cooling at a rate of about 50°C/hour.(39) There was essentially no increase for a full-annealing cooling rate, e.g., less than 1°C/hour.

It has not been established what cooling cycle will be used; BNWL indicates that cooling probably would be in the range from slow cooling to free air convection, which might result in an increase of about seven times in surface area. Whether this relative surface area increase is the same for the larger-diameter cylinder, e.g., 30 cm (12 in) is not known. Because of the inherently slower cool-down rate, the larger-diameter cylinder would probably have a somewhat lower relative surface area increase for a given mode of cooling. For a slow-cool or annealed case, a soaking pit operation would be required and such a "storage" operation would probably add to the problems of facility throughput.

In the as-cast condition, a 0.3 m (12 in) diameter by 3.0 m (10 ft) high cylinder, has a surface-to-mass ratio of about 0.05 cm^2/g. For reasonably slow cooling, a multiplier of seven has been assumed, so the surface-to-mass ratio would be 0.35 cm^2/g.

Another factor that must be considered in setting the surface-to-mass ratio is that of possible mechanical damage to the glass of the cylinders. Very high impact velocities, e.g., 21 m/sec (70 ft/sec) can increase glass surface by a factor of perhaps ten or more. On the other hand, fairly nominal impacts, e.g., 1.5 m/sec (5 ft/sec), would not increase the overall surface area by more than about 10%. There is no reason to assume that any canisters subjected to a high-impact force would be sent to disposal; hence, the nominal kind of incident is the type that should be considered. A 10% increase of the surface-to-mass ratio, as developed above from the cooling analysis, would increase the surface to about 0.4 cm^2/g. This surface-to-mass ratio is equivalent to that of a cube of glass 4.7 cm on an edge containing approximately 332 g of glass. This initial "particle size" has been used in the bulk leaching analysis presented below in Section B-3.6.1.3.5.

B-3.6.1.3.2.2 Devitrification

Although there has been a great deal of discussion of possible devitrification of glass, there is no basis at the present time to assume that such devitrification will occur. Tests have been run on various high-level waste glasses, at different temperatures, "spiked" with curium in order to develop radiation damage effects more rapidly. Samples with curium were prepared in 1974 and these samples have developed an equivalent age of up to 2000 years from the standpoint of radiation damage.(41) Many characteristics have been measured, such as stored energy, density change, leach rate, physical strength, etc. Relatively modest changes in leach rates have been noted, not more than a factor of two higher, but the radiation exposure does not seem to have any bearing on devitrification. Similar results were obtained by the Karlsruhe Laboratories in Germany. The studies indicate that devitrification processes are basically effects caused by high temperatures, generally in the range of 700-900°C, but some devitrification may occur in the 400-500°C range.

Devitrification, if it were to occur, might increase the leach rate by a factor of approximately ten. Specifically, effects of various devitrification temperatures observed by Ross(39) indicate that the bulk leach rate increased by a factor of six with the peak occurring at a devitrification temperature of 700°C.

B-3.6.1.3.3 Calcine Leach Rates

Calcine leach rates are generally high, with values reported of about 0.1 g/cm^2-day.(11) This rate would result in total dissolution of a calcine, even assuming that it had a surface-to-mass ratio as low as that of the bulk glass described above, in less than 60 days. For all practical purposes, then, calcine dissolution _per se_ does not represent a delay function in the waste disposal release scenario.

B-3.6.1.3.4 Spent Fuel Dissolution Rates

The major development work on the leaching of spent fuel was performed by BNWL.(42) This study utilized particles of high-burnup light water reactor fuel pellets and attempted the dissolution of these fuel particles with Hanford ground water, distilled water, and deionized water. Leaching of chemical elements (curium, plutonium, strontium, and cesium) was evaluated. Within the leach time of those studies, 150 days, the leach rate constants that were established ranged from about 4 x 10^{-9} g/cm^2-day to about 7 x 10^{-6} g/cm^2-day. The lowest values were for dissolution derived from curium removal measurements while the high rates were based on cesium.

The data reported further indicate that after one day, the "leach" values are on the order of 100 times higher than after 150 days. These data indicate that fuel assembly leaching might be less than that of high-level waste glasses, certainly not the anticipated result.

In order to estimate the long-term leachability of spent fuel, it is necessary to consider whether conditions may be present that will cause the fuel to oxidize, i.e., to change from its initial ceramic form to a simple, finely-divided, oxide mixture. If oxygen (air) is available to spent fuel oxide pellets at temperatures much above 120-150°C (250-300°F), the fuel rapidly oxidizes from UO_2 to U_3O_8, and in so doing, forms a finely-divided powder that expands to the point where fuel cladding is ruptured. The effect would propagate from the first small pinhole to develop in the Zircaloy cladding. If this phenomenon occurred, leaching of the fuel would be at least as rapid as with calcine. How much effect water of varying chemistries would have on a dissolution rate of fuel is not known, and therefore a reasonable single reference value for spent fuel leaching cannot be set with any confidence. The leaching values that can be hypothesized, based on non-oxidizing conditions, could be similar to those of the high-level waste glasses.

At the other extreme, if hot, oxidizing conditions are assumed, leach rates could well resemble those of calcines, i.e., rapid.

B-3.6.1.3.5 Total Leach Times

B-3.6.1.3.5.1 Glass Leach Times

In order to establish the quantity of material that might be leached under idealized situations for subsequent transport to the environment, the conditions previously reviewed have been integrated into an overall removal function.

For a cube initially of side x_o cm, with density ρ g/cm^3, with a specific leach rate constant of k g/cm^2-day, it is easy to show that after a time of Θ days, the side of the cube has shrunk to x cm, where

$$(x_o - x) = (2k/\rho)\,\Theta \tag{1}$$

at this point the weight is $w = \rho\, x^3$, as compared with the initial weight $w_o = \rho\, x_o^3$. The above is derived by integration of the dissolution rate at Θ days after the start of leaching:

$$-\frac{dw}{d\Theta} = 6kx^2 \tag{2}$$

Using these relations with the values already given (see Section B-3.6.1.3.2.1) for the unit glass cube: $x_o = 4.7$ cm, $\rho = 3.2$ g/cm^3, and $k = 10^{-6}$ g/cm^2-day, one can calculate the curves shown in Figures B-41 and B-42.

Figure B-41 shows the percent of the glass remaining (unleached) vs time, and Figure B-42 shows the leach rate in percent of the original (total) glass being leached each year. Note that with a conservative assumption, it will all be dissolved in 20,000 years, one-half of it being dissolved in about 4000 years.

Figures B-43 and B-44 show the analogous results, assuming the more optimistic, but quite possibly attainable, leach rate constant of 10^{-8} g/cm^2-day. Under this assumption, the glass would last two million years, one-half of it being dissolved in the first 400,000 years. The values shown are relatively conservative, allowing for the fact that leach rate, particle size, water chemistry, and other parameters are unknown and cannot be projected with any certainty to perhaps hundreds of thousands of years into the future.

B-3.6.1.3.5.2 Calcine Leach Times

Simple calcines are rapidly soluble, the usually-reported leach rate value being 10^{-1} g/cm^2-day. Furthermore, calcines are usually finely divided; therefore, full dissolution can be calculated to occur in a few days to a few months. This rapid dissolution rate indicates that

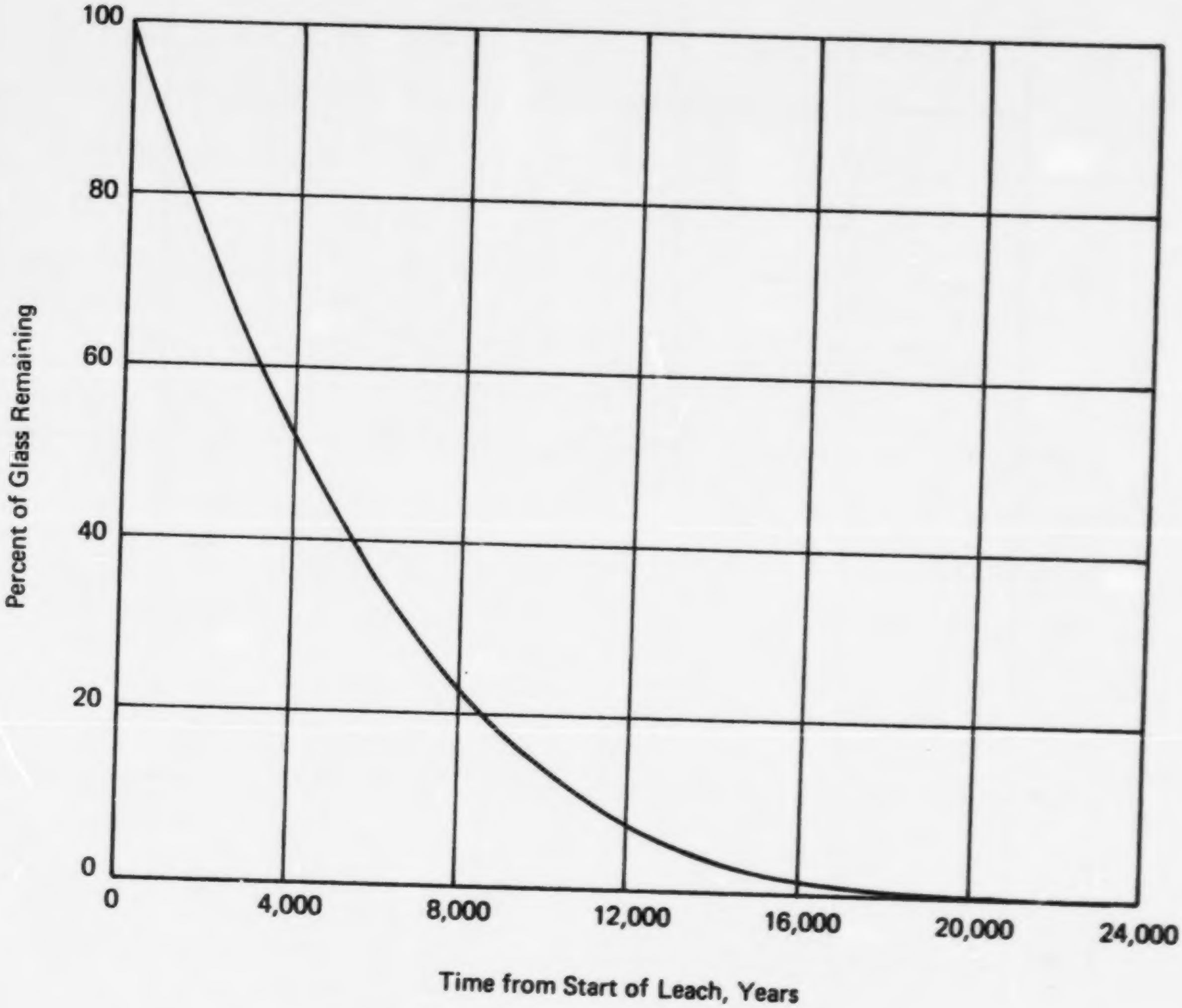


FIGURE B–41 PERCENT OF GLASS REMAINING (UNLEACHED) VS. TIME $k = 10^{-6}$ g/cm^2-day (CONSERVATIVE VALUE)

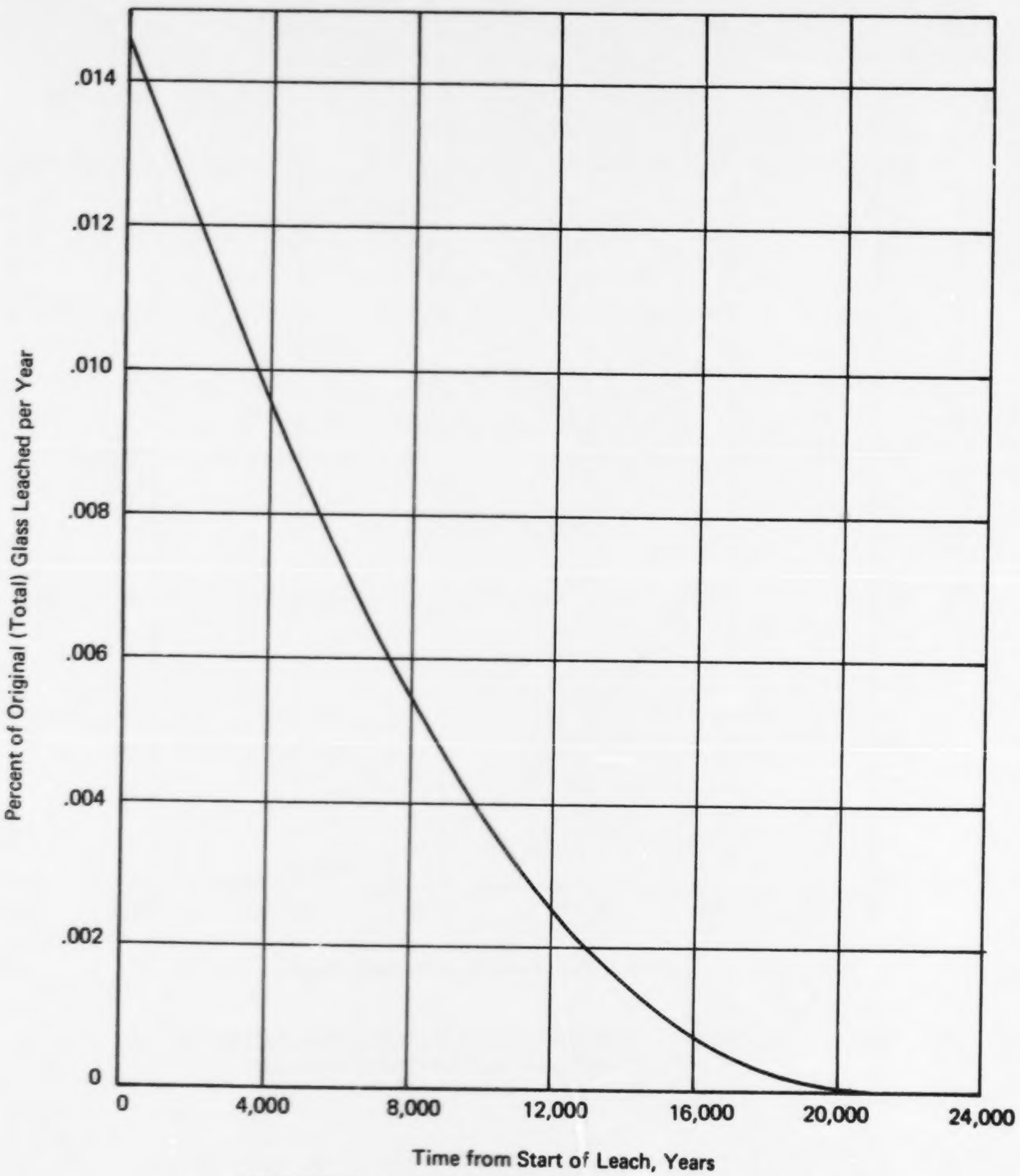


FIGURE B–42 ANNUAL LEACH RATE VS. TIME, k = 10^{-6} g/cm^2-day (CONSERVATIVE VALUE)

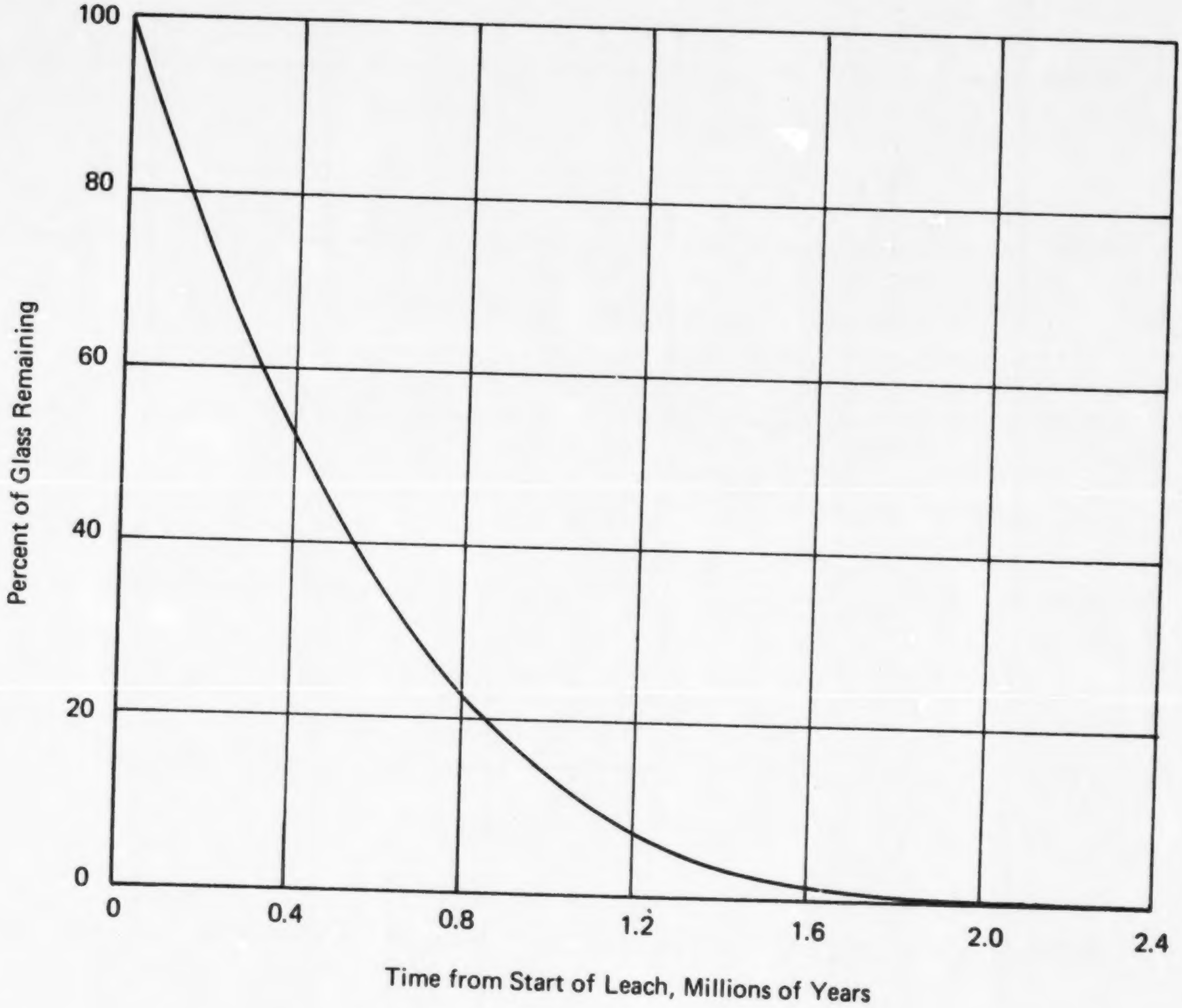


FIGURE B–43 PERCENT OF GLASS REMAINING (UNLEACHED) VS. TIME
$k = 10^{-8}$ g/cm^2-day (POSSIBLY ATTAINABLE VALUE)

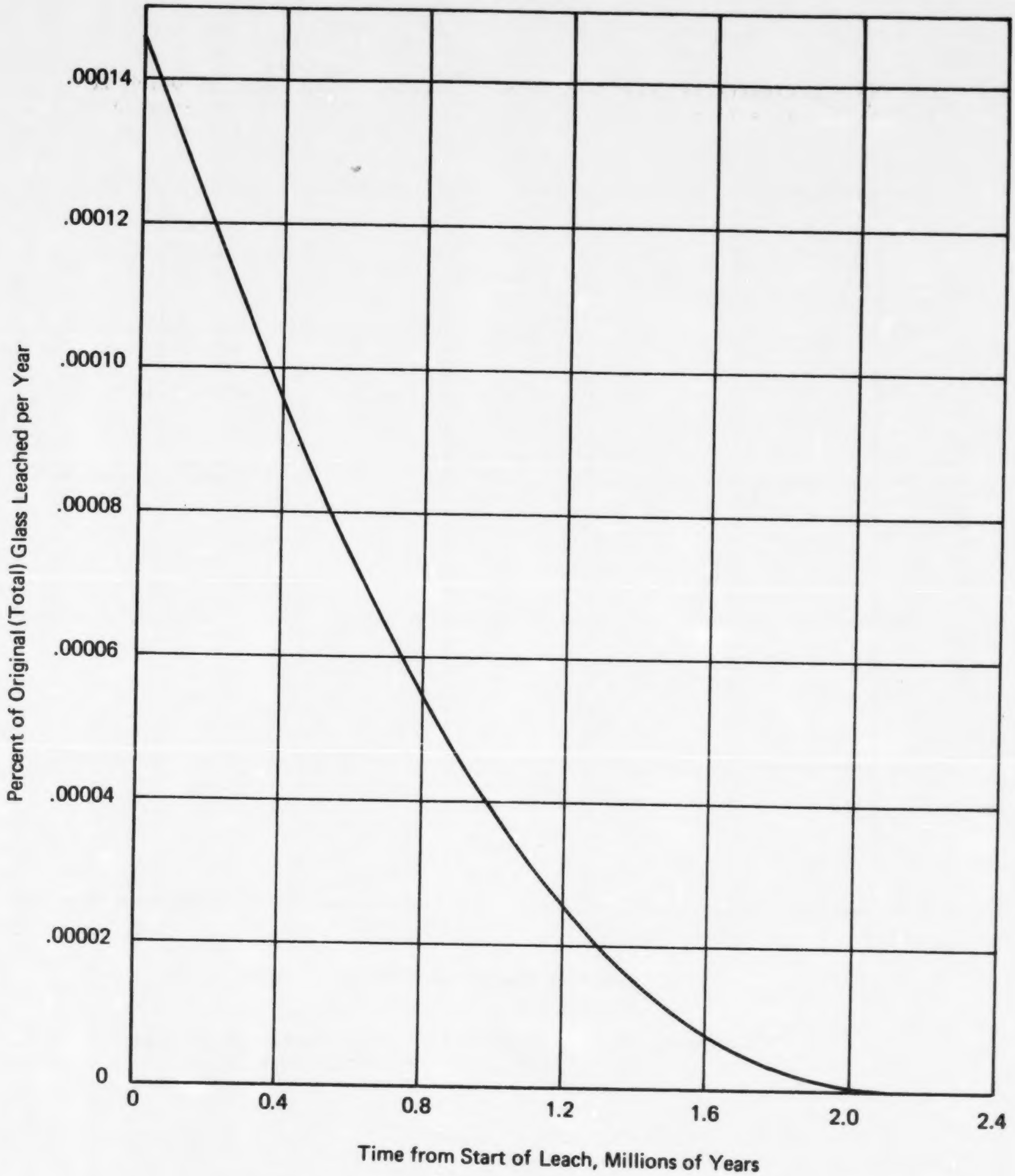


FIGURE B–44 ANNUAL LEACH RATE VS. TIME, k = 10^{-8} g/cm^2-day (POSSIBLY ATTAINABLE VALUE) calcine containment is dependent primarily upon the canister in which it is buried. In a salt repository, there is not much difference between the corrosion resistance of carbon or stainless steel. Carbon steel has perhaps a more uniform corrosion rate, but stainless steel is more subject to stress chloride cracking. Although the mechanisms are different, both might be breached in a relatively short time. The failure point is probably about 50 years; if the geologic phenomenon that could start the leaching process occurs, the material is effectively available for leaching and transport in a short period of time.

It would be desirable, if at all feasible, for the canister to provide some longer-term retention for a material like a calcine, which has little integrity of its own. About 500-1000 years would be a suitable period, since in this time frame the fission products are controlling and are generally more soluble than the TRUs.

Titanium might be useful as a container for calcine in a salt repository. The relatively standard titanium alloys have a corrosion rate of about 0.025 mm (1 mil) per year, for a high temperature salt wet/dry situation (see Appendix B-V). Some alloys of titanium, including one with palladium, would have much better corrosion properties, but would be substantially more expensive. It appears that 0.025 mm (1 mil) per year would be acceptable as a corrosion rate; therefore, using "commercial" material and thickening the wall appropriately to last 500-1000 years would be a better approach than using a more expensive canister. This approach should be explored to establish the optimum cost/benefit. For the purpose of estimating hazards, a canister lifetime of about 800 years could be assumed, but after canister failure, fairly rapid release of the waste calcines must be assumed.

A "supercalcine" might be developed for most waste compositions that would have very good leach resistance characteristics, but it appears that the supercalcines must be specifically tailored to a given waste composition requiring a degree of control not necessary for glasses. Furthermore, because the procedures for tailoring and sintering are not much different from those of glass production, there appears to be no significant advantage to this approach.

B-3.6.1.3.5.3 Spent Fuel Assembly Leach Times

The containment in series is first, the canister; second, the zirconium fuel cladding; and third, the ceramic UO_2. As indicated in the prior discussion, the leach rate applicable to spent fuel assemblies is highly speculative. Data in BNWL-2057(42) suggest that spent fuel assemblies can be assumed to be similar in leach characteristics to the glass previously discussed. Depending on the oxygen content of the ground water, however, the UO_2 in a heated fuel assembly may be oxidized to the U_3O_8 state. If this were the case, it would have leaching characteristics similar to that of the HLW calcine. In this case, the short-term integrity could be determined, similar to that of the calcine, by the canister lifetime characteristics. Depending on the temperature and the oxidizing nature of the ground water, a spent fuel assembly may be assumed to have leach characteristics as low as those of glass or as high as those of calcine.

B-3.6.1.4 Solubility Considerations

The leaching rate data given above will result, for any given ratio of leachant flow to high-level waste weight, in a particular set of concentrations of all soluble species. For low flows, however, the very low solubilities of the actinide compounds may limit their concentration in the leachate.

Rai has correlated the solubilities of the actinide compounds.(40) His approach was to determine the most insoluble of all the feasible compounds for each actinide, then to calculate from solubility products the concentration of all soluble actinide-bearing ions in equilibrium with that compound. These solubilities are a function of pH, since H^+ ion is usually involved in the equilibrium. Thus, in the dissolution of PuO_2 to form PuO_2^{++} ion,

$$PuO_2 + 2H^+ + 1/2\ O_2 = PuO_2^{++} + H_2O \qquad (3)$$

the equilibrium PuO_2^{++} concentration (solubility) ought to increase with the square of the H^+ ion concentration, or the log (PuO_2^{++}) ought to decrease in accordance with twice the pH.

Solubility relations such as this are summarized in Table B-21 for the four principal actinides, based on curves in BNWL-1983.(40) Relationships are given in saturated salt solutions and in non-salt solutions like those found in rock.

When leach calculations are made, if the actinide concentrations in the leachate should exceed the values calculated from Table B-21 , they should be reduced to those values.

B-3.6.2 Reference Technologies in Other Media

In the absence of published information, it can only be assumed that the same leach rates presented above for the salt environment would apply roughly in a rock repository as well. The actinide solubilities would be different, however, and the differences have been shown in Table B-21.

TABLE B-21
SOLUBILITY LIMITS OF PRINCIPAL SPECIES FROM ACTINIDE DISSOLUTION

	In Sat'd NaCl Solution		In Water in Rock	
Actinide	Least Soluble Solid	Most Soluble Species in Solution	Least Soluble Solid	Most Soluble Species in Solution
U	Na_2UO_4	log $[UO_2Cl^+]$= -0.7-(4)(pH)	$UO_2(OH)_2$	log $[UO_2^{++}]$=5 -(2)(pH) w/CO_2 @ .0003 atm. in alkaline medium, log $[UO_2^{++}]$ < 11.2 -(2)(pH)
Np	$NpO_2(OH)_2$	log $[NpO_2^+]$ = 4-pH	$NpO_2(OH)_2$	log $[NpO_2^+]$ = 4 - pH w/CO_2, same
Pu	PuO_2	log $[PuO_2(OH)°_2]$ = -11.5 log $[PuO_2Cl°]$ = -4.8-pH	PuO_2	log $[PuO_2(OH)°_2]$ = -11.5 log $[PuO_2^{++}]$ = -0.5-(2)(pH) log $[PuO_2^+]$ = -5.3 - pH w/CO_2, above plus log $[PuO_2(CO_3)(OH)_2^{=}]$= -27.2+(2)(pH)
Am	AmO_2OH	log $[AmOH^{++}]$ = -4.4 -(2)(pH) log $[AmCl^{++}]$ = 3.3 -(3)(pH)	AmO_2OH	log $[AmOH^{++}]$ = -4.4 - (2)(pH) Effect of CO_2 unknown

Note: All logarithms are to the base 10. Solubilities in g mols/liter

Source: Based on data in Ames, L.L., et al, "A Review of Actinide-Sediment Reactions...," BNWL-1983, Battelle Northwest Laboratory, February 10, 1976.

B-4.0 COST CONSIDERATIONS

B-4.1 GENERAL

The purpose of this section of the report is to present data on the comparative costs of the various reference cases discussed in Section 3.0. Such information will be needed in order to evaluate the cost effectiveness of alternative disposal techniques. Since only limited amounts of cost data have been published, and because it was beyond the scope of this study to do independent cost-estimating, the data presented should be considered only as rough estimates. Nevertheless, they provide useful perspective on the relative economics of various disposal techniques.

Two basic types of HLW are considered in this section--solidified HLLW from reprocessing operations and spent fuel elements from a throwaway cycle. Both types are assumed to be suitably packaged for nonretrievable disposal in deep geologic formations (repositories). The effect (on the cost) of incorporating retrievability into the design of such a repository will also be shown.

The two waste paths considered are:

For Disposal as Solid HLW	For Disposal as Spent Fuel
• Storage of spent fuel at reactor sites and at interim storage sites for a total of 10 years.	• Storage of spent fuel at reactor sites and interim storage sites for a total of 10 years.
• Transfer of spent fuel to reprocessor.	NA[*]
• Reprocessing	NA
• Prompt conversion of HLLW to canned solid HLW.	NA
• Transfer of HLW canister to repository.	• Transport of spent fuel to repository.
NA	• Canning of spent fuel.
• Burial of HLW canisters.	• Burial of spent fuel canisters.

Thus, the sequence for the solidified HLW disposal scheme is what has generally been assumed in the past (but with delayed reprocessing), with prompt conversion of the reprocessing wastes to canned solids, for disposal at a Federal repository. The sequence for the spent-fuel disposal scheme (the "throwaway cycle") assumes the identical combined ten-year storage of spent fuel at the reactor site and at a centralized

---

[*] Not Applicable

Preceding page blank

interim storage facility, and finally burial at the repository. The spent-fuel transport industry has evolved on the principle of shipping uncanned fuel in special casks; the same approach would probably be used to transport spent fuel from interim storage facilities to the repository, where final canning would take place. Specialized equipment and machinery are required with stringent quality control; therefore, it seems more reliable and more cost-effective to perform canning at a single site (the repository) rather than at multiple interim storage sites or reactor sites.

For each approach, approximate cost figures will be given, with the exception of the following items, which are beyond the scope of this assignment:

- Reprocessing costs will not be given.
- Transportation costs will be estimated as a general range only.

B-4.2 SPENT-FUEL STORAGE COSTS

Storage of spent fuel at reactor sites (using high-density storage racks in lieu of the normal-density racks hitherto used) and at interim facilities for a total of ten years can be performed for costs of about \$80-150/kg of heavy metal (HM)*, depending on the financing costs and including an allowance for intermediate shipment at \$10-20/kg HM. These costs are the same, regardless of which of the two waste paths is chosen.

B-4.3 POST STORAGE/PRE-BURIAL COSTS

B-4.3.1 Solid HLW Disposal Path

B-4.3.1.1 Transportation Costs (to Reprocessor/Solidification Plant)

Transportation of ten-year-old spent fuel from interim pool storage to the reprocessing site would cost about \$8-18/kg HM. No cost is assigned in this study to the reprocessing itself. Because the solidification facility is assumed to be at the reprocessing site, there is no transport charge for HLLW movement.

B-4.3.1.2 HLLW Solidification Costs

The solidification process will probably take place at the reprocessing facility. Although it is possible that the reprocessing facilities will also be located at, or adjacent to, the HLW repository, location is a factor reflected only in charges for shipping solidified waste, which will be about the same for all alternatives (see below).

---

*Heavy metal means the uranium and plutonium in the fuel originally charged to the reactor.

The HLW solidification facilities are assumed to handle the normal throughput of a reprocessing plant, which is in the range of 4-8 MTHM/day. A 5 MTHM/day (or 1500 MTHM/yr) size is chosen to be the same as that of the AGNS facility and that proposed by Exxon.

In considering the relative costs of the competing solidification processes, it should first be noted that capital costs dominate and equipment cost is a relatively small fraction of the total investment. The major portion of the capital investment is for the "hot cell" itself plus associated auxiliary support systems, e.g., remote handling and maintenance equipment, the off-gas treatment system, etc.

One of the key determinants of hot-cell costs is physical size, especially area. Area depends upon the space needed for process equipment and is particularly sensitive to the amount of redundancy required for such equipment. Thus, if one of the processes were judged to require inherently more redundancy or a substantially greater cell area for a given throughput, that process would clearly have an economic disadvantage relative to other processes with more compact layouts. In gauging the amount of redundancy required, particular attention must be paid to reliability and maintenance problems in a highly radioactive environment.

To date, only one detailed study of solidification costs has been published.(43) Cost data on total reprocessing plants have been reported, however, and an attempt can be made to allocate a portion of total costs to the HLLW solidification functions.

Data have been published for two reprocessing plants involving HLLW solidification--the proposed Exxon Nuclear Fuel Recycle Facility and the AGNS plant. In addition, the S. M. Stoller Corporation (SMSC) and Battelle Northwest Laboratories (BNWL) have independently developed such cost data.

B-4.3.1.2.1 Exxon Nuclear Fuel Recycle Facility

The Exxon Fuel Recycle Facility PSAR(44) indicates that the concentrated HLLW will be sent to a fluidized-bed waste calciner utilizing aluminum oxide as a starting granular material and with a fluidizing gas of superheated steam. The calcined wastes then flow to a mixing/metering vessel, where they may be combined with an added material suitable for glass-forming or fed directly to a waste-melting furnace. The melt then flows into waste canisters that are air-cooled and are then temporarily stored in a water pool beneath the process operating cell.

In the published data Exxon has not allocated costs to this calcining/glassification operation, but has estimated the total Recycle Reprocessing Facility capital cost at approximately $600 million(45) in 1975 dollars. This includes, of course, all of the common auxiliary service functions required for the operation of the entire facility--

spent fuel assembly storage, cooling towers, fire water system, roads, security, chemical tank farms, sanitary wastes, emergency power, ventilation system, administration facilities, etc.

In order to estimate roughly the fraction of capital costs that should be allocated to the HLW solidification portion of the plant, the various process cell areas in the facility have been analyzed; the HLW solidification cell constitutes approximately 5% of the total remote and contact cell area of the Exxon plant. If 60% of total facility cost is directly associated with the reprocessing building itself, the HLW solidification fraction would be 3% of the total project cost. If a 10% per year escalation is assumed since 1975 to correct for inflation, the 1977 direct capital figure for the waste solidification cell would be about $23 million. An additional allowance must be included for interest during construction, which if calculated at 8% per year would add somewhat more than $6 million, for a total rounded capital cost of $30 million (1977 dollars) for waste solidification. The equivalent unit cost, at an annual fixed charge rate of 20%, is approximately $4/kg HM. Process changes may result in somewhat more or less space required in the reprocessing plant, but the resultant cost impact should be relatively small. Changes attributable to equipment cost variations are expected to be even smaller and relatively unimportant as compared with the cell cost elements.

B-4.3.1.2.2 Allied General Nuclear Services

The AGNS plant at Barnwell, S.C. does not currently have a waste-solidification step, because regulatory criteria were not defined when the plant was constructed. The HLLW were to be concentrated and then stored in stainless-steel tanks pending resolution of regulatory criteria on solidification processes and facility design. AGNS has announced(46) total costs for upgrading the facility to include waste solidification as well as other steps. The published data are insufficient to partition the costs among the various additional process steps, however.

B-4.3.1.2.3 Western Reprocessing Study

In a preliminary cost analysis for the Western Reprocessing Study Group, SMSC developed a value of approximately $120-130 million of total waste treatment facilities associated with a reprocessing plant costing approximately $600-650 million in 1976 dollars. The waste treatment facilities included not only HLLW solidification, but also HLLW concentration and storage, as well as the cost of all other waste treatment facilities in the plant, e.g., facilities for treatment of intermediate- and low-level TRU wastes, etc. It is estimated that on the order of one-quarter to one-third of total waste-treatment costs would be allocated to the HLW solidification step, or $30-45 million, a value roughly comparable to that inferred above from published data on the Exxon facility.

Another item of information useful for rough comparisons is the estimated cost of the HLW calcining facility at Idaho Falls for defense wastes, approximately \$65 million.(47) Although this facility is not the same as one for solidification of commercial HLW, many of the features are similar, e.g., off-gas treatment systems, support systems, etc.

B-4.3.1.2.4 Battelle Study

In BNWL-1667,(43) a cost optimization was performed to determine the sensitivity of waste treatment costs to variables of waste type, cooling time, and solidification process. The calculational code was based on a 1370 MTU/yr reprocessing throughput, 20-year plant life, a 30/70 debt/equity ratio, 15% return on equity, 8% interest on debt, and included adjustments for equipment depreciation and tax structure. The code calculates cost sensitivity to varying storage times, given a waste type and solidification process.

The BNWL-1667 computational code gives results in mills/MWh, which may not be directly comparable to other results expressed in \$/kg, or total capital costs, because the relations between mills/MWh, \$/kg, and capital costs are a function of the economic assumptions used for different studies; nevertheless, certain observations may be made regarding the results.

The BNWL-1667 data confirm that the waste solidification process selected (assuming equivalent reliability) has very little effect on total costs, although longer cooling times will decrease solidification costs of any one system. Total costs of storage-plus-solidification (including canning) for five-year-old waste vary from 28 to 36 mills/MWh (1972 dollars) over a range of waste solidification techniques from pot calcination to spray melter solidification. Note that 28 to 36 mills/MWh corresponds to about \$7.4-9.5/kg, or, escalated to 1977 at 10% per year, about \$12-15/kg.

Based on the foregoing discussion, the cost of waste solidification facilities should fall in the range of \$30-60 million, or, \$4-15/kg HM. Cost variations due to alternative calcining or glassification processes are likely to be small (\$1-2/kg HM) and well within the uncertainty band of these rough estimates of total waste solidification costs.

B-4.3.1.3 Canister Costs

Waste canister costs vary with the material and thickness chosen, with carbon steel at \$1/kg HM, stainless steel at \$2/kg HM, and titanium at \$4-6/kg HM.

B-4.3.1.4 Transportation Costs (to Repository)

Shipping costs are not expected to differ greatly between calcine or glass, and both ought to be about one-quarter to one-third of the cost for shipping the equivalent amount of recently-discharged spent fuel (allowing for the higher density of waste permissible per unit volume, since it is ten years old). This leads to a solidified-waste transportation cost of about \$3-8/kg of original HM (not per kg of solidified waste). The low end of the range is, of course, more likely if the reprocessing unit is located at or near the repository.

B-4.3.1.5 HLW Solidification Cost Conclusions

It is very difficult to compare different cost estimate sources directly without a detailed comparison of cash flow assumptions. Thus, the value of the cost data presented in this report lies not so much in the absolute cost figures given, but rather in the following general conclusions:

- Costs are largely insensitive to waste solidification methods, assuming equivalent reliability.
- Cooler wastes are less expensive to treat, ship, and dispose of, but these cost savings are offset by added storage costs.

B-4.3.2 Spent-Fuel Disposal Path

B-4.3.2.1 Transportation Costs (to Repository)

Transportation of ten-year-old spent fuel from interim pool storage to the repository is assumed to be \$8-18/kg HM, the same as in Section 4.3.1.1.

B-4.3.2.2 Canning of Spent Fuel

It is assumed that the fuel will be placed in gas-filled, sealed outer canisters in a canning facility at or adjacent to the repository. The major components of the spent-fuel canning cost are:

1. Fabricated canister component costs.
2. Canning facility fixed charges - the capital charges associated with the loading, sealing, and testing facilities necessary for the canning process.
3. Canning facility operating and maintenance charges.

The fabricated canister components are estimated from \$2/kg HM for carbon steel, to \$6/kg HM for stainless steel, to \$15/kg HM for titanium.

Capital costs for a canning facility have not been published to date. Based on the number of operating locations in a hot cell, such a facility might be built to handle 3000 MTHM/yr at a cost of roughly \$100 million. Allowing for possible variations in this figure and in facility lifetime, the equivalent unit cost is in the range of \$5-10/kg HM.

Operation and maintenance charges for such a facility keyed to a staff of about 150, are estimated to add another \$1-2/kg of HM.

When the above cost ranges are combined, the resulting range of total costs is \$8-27/kg HM; most of this range of variation is due to the \$13/kg HM variation in the estimated costs of fabricated canisters.

B-4.4 REPOSITORY COSTS

B-4.4.1 Costs for Geologic Disposal in Salt

Order-of-magnitude cost estimates for geologic disposal in bedded salt have been developed for the reference facilities discussed in Section 3.5 of this report. The estimates (in 1977 dollars) for both retrievable and non-retrievable facilities, designed to receive either canistered HLW from a reprocessing cycle, or canistered spent fuel from a throwaway cycle are summarized in Tables B-22 and B-23. The estimated costs for mine excavation, waste handling, equipment design and fabrication, and overall construction costs have no allowance for contingencies.

Mining costs are based on a review of recent bid data for salt dome production in Louisiana, as well as on recent cost experience in underground mine construction. The mine general arrangement, as discussed in Section 3.5, is similar to that evolved for OWI.(17) Each of the four shafts is concrete-lined and sunk to a depth of 460m (1500 feet) through hard rock (granite). The mine excavation costs cover all costs for mining-related activities over the life of the facility, including surface support facilities, mining machines, underground and aboveground materials handling and hauling equipment, and mining manpower. Thus, whether all mining is done prior to facility operation or concurrent with facility operation, the overall mining cost component will not change. (It should be noted that normal commercial mining accounting practices would probably consider much of the mine excavation as an operating expense rather than a capital item.) A unit excavation cost of \$65/m^3 (\$50/yd^3) for salt has been assumed.

Unit costs for canister holes have also been developed, based on conventional mining data and the conceptual canister hole designs discussed in Appendix B-IV. For non-retrievable storage, costs for HLW are \$1000 to bore the hole, insert the waste, and backfill the hole.

TABLE B-22

APPROXIMATE COSTS REFERENCE REPOSITORY IN BEDDED SALT - NON-RETRIEVABLE STORAGE FOR EQUIVALENT OF 107,000 MTHM

	Millions of (1977) Dollars	
Cost Component	HLW	"Throwaway" Case
Surface facility	125	125
In-mine structures	60	60
Excavation		
Low-level TRU	85	50
Intermediate-level TRU/ Cladding Hulls	314	127
HLW/Spent fuel	503	1572
Excavation Sub-Total	902	1749
Shaft costs	18	18
Canister holes		
HLW	36	494
Intermediate-level TRU	314	163
Facility backfill	30	58
Facility decommissioning	10	10
Total Capital	1495	2677
Average Annual Operating and Maintenance Costs (over 10-year operating period)	30/yr	30/yr
Total (Undiscounted) Cost for Capital + 10 years of operation	1795	2977
Total (Undiscounted)	$17/kg HM	$28/kg HM

TABLE B-23

APPROXIMATE COSTS REFERENCE REPOSITORY IN BEDDED SALT - FULLY RETRIEVABLE STORAGE FOR EQUIVALENT OF 107,000 MTHM

	Millions of (1977) Dollars	
Cost Component	HLW	"Throwaway" Case
Surface facility	125	125
In-mine structure	60	60
Excavation		
Low-level TRU	85	50
Intermediate-level TRU/ Cladding hulls	314	127
HLW/Spent fuel	503	1572
Excavation Sub-Total	902	1749
Shaft Costs	18	18
Canister holes		
HLW	186	1728
Intermediate-level TRU	282	117
Facility backfill	451	875
Facility decommissioning	10	10
Total Capital	2034	4682
Average Annual Operating and Maintenance Costs (over 10-year operating period)	30/yr	30/yr
Total (Undiscounted) Cost for Capital + 10 years of operation	2334	4982
Total (Undiscounted)	$22/kg HM	$46/kg HM

For retrievable storage, costs for HLW are about $5000 to bore the hole, provide a one-inch thick carbon steel canister liner and concrete shield plug, install the liner, and place the waste canister and hole plug.

The facility design is still in the conceptual stage. Furthermore, the NRC has not yet issued any design criteria directly applicable to waste isolation facility design. It was therefore necessary to draw heavily on experience in other nuclear facilities when developing cost estimates for the reference waste burial facility structures and equipment. The basic criterion in sizing the surface receiving facility and the in-mine canister-handling stations is that the facility can physically handle the required throughput of 350,000 canisters over the assumed operating lifetime of ten years.

Necessary plant structures are discussed in Appendix B-IV. Briefly, however, in-mine structures include the HLW and intermediate-level TRU receiving/transfer cells and the man and materials receiving station. Surface facilities include a canistered waste receiving building, canistered waste transfer cells, an interim canistered waste storage building, a canistered waste hoist cell and hoisting equipment, a low-level TRU receiving building, an interim TRU storage building, a TRU hoist facility, a man and materials hoist facility, a mine ventilation system building, a site radwaste process facility and plant stack, and site support facilities, such as an administration building, machine shop, warehouse, security gate houses, and a general personnel staging facility containing health physics and chemistry laboratories, a lunch room, showers, lockers, etc. Nuclear facility and equipment cost data from WASH-1230(48) escalated to 1977 dollars, were used where applicable. It was arbitrarily assumed for costing purposes that the plant radwaste and ventilation systems, the interim canistered waste storage area, the canistered waste handling cells, and the canistered hoist cell and hoisting equipment will all be housed in the equivalent of seismic Category 1 structures. Construction cost data were also taken from R. S. Means(49) where applicable. Overall structure and equipment requirements are similar for each of the respective reference facilities in Tables B-24 and B-25. Annual operating and maintenance costs include staff salaries and benefits (for all staff, except those associated with the mining operation), facility maintenance, and costs for electricity, water, fuel, etc. Facility staff requirements are summarized in Table B-24. Because canister handling is assumed to be a continuous 24-hour, 7-day operation, a rotating 4-shift operation is necessary for personnel associated with canister handling (equipment operators, mechanics, health physics support, and a skeleton maintenance crew). Security will also operate on a rotating 4-shift basis.

For purposes of comparison of repository costs with other waste disposal costs, a unit cost value ($/kg HM charged) has been included for each reference facility in Tables B-22 and B-23. This number is derived by summing the total facility capital costs and ten years

TABLE-24

PLANT STAFF ESTIMATE FOR REFERENCE REPOSITORY

		Total Staff
SURFACE FACILITY		
General Staff		
Administrative and Clerical		10
Security		21
Chemistry		5
Health Physics		21
Maintenance		19
Custodial		4
	Sub-Total	80
Canistered Waste Handling		
Yard Crew		20
Cask Handling		136
Hot Cell Operation		36
	Sub-Total	192
Low-level TRU Handling		3
Transfer Hoist Operators		13
Radwaste/Laundry		4
	Surface Facility Total	292
BELOW GRADE		
Low-level TRU Handling		5
Canistered Waste Receiving Facilities		24
Canistered Waste Transport and Burial		240
Underground Maintenance		12
	BELOW GRADE LEVEL	281
	FACILITY TOTALS - Surface Facility	292
	Below Grade	281
	TOTAL	573

TABLE B-25

SUMMARY OF HIGH-LEVEL WASTE DISPOSAL COSTS

(In Salt, Non-Retrievable)

	$/kg HM (1977 Dollars)	
	HLW Case	"Throwaway" Case
Early storage of spent fuel (10 yr)	80-150	80-150
Transport to reprocessor	8- 18	-
(Reprocessing)	(Not considered)	-
Solidification of HLLW, inc. canning	6- 23	-
Transport to repository	3- 8	8- 18
Canning	(Incl. in Solidif'n)	8- 27
Burial of canisters	17*	28*
Total	114-216	124-223

---

*Retrievability would add 30-60% to these values.

of operating costs (assumed active operating design life) and dividing by the total facility capacity (107,000 MTHM charged). While this is a simplified cost treatment, it is adequate for present purposes.

B-4.4.2 Cost Versus Depth

Cost versus depth was not examined in detail for this study, but it may be assumed that deeper burial will be more costly. Shaft costs will be higher; mining costs will be higher, because of the longer time to transfer men and materials into and out of the mine; mine operating costs will be higher for the same reasons. Stability criteria for mines at greater depth may also have an adverse impact on the general arrangement of the mine and on maximum permissible planar heat density.

B-4.5 COST SUMMARY FOR HLW OR SPENT FUEL DISPOSAL IN SALT

The various elements contributing to total waste disposal costs are summarized in Table B-25. The cost is dominated by the interim storage costs. For comparison, total fuel cycle costs might be roughly \$1000/kg HM for a prompt reprocessing cycle. Hence, interim storage alone can increase these costs by 8-15% depending upon the type of storage facility.

B-4.6 COSTS FOR GEOLOGIC DISPOSAL IN HARD ROCK

Detailed cost estimates could not be developed for geologic disposal in hard rock since suitable planar heat loading design criteria have yet to be established for hard rock. Unit costs for excavation have been examined and, based on preliminary findings, are expected to be 30-100% higher for hard rock than for salt at the same elevation. Preliminary indications are that hard rock heat input might be limited to something on the order of 50% that of salt.(30,50) In that case, overall excavation costs in hard rock would be on the order of 2.6-4 times as high as for salt at the 460-meter mine depth assumed for this study, and that overall facility costs for hard rock would be two to three times as high as those for salt for the same waste stored.

B-4.7 NOTE ON THE DISPOSAL COSTS FOR LOW-LEVEL TRU WASTES

Table B-26 shows estimates of costs expected for a geologic disposal facility handling 140,000 m^3 of low-level transuranic (TRU) wastes. The surface and handling facilities will be much less expensive than for HLW because shielding is not required. Excavation costs per unit of waste volume will be much less than for HLW because thermal loading restrictions will not be limiting. With lower radiation levels, annual operating costs are lower.

TABLE B-26

REFERENCE FACILITY COSTS (BEDDED SALT) - NON-RETRIEVABLE STORAGE OF LOW-LEVEL TRU WASTE

Cost Component	Millions of (1977) Dollars Non-Retrievable	Millions of (1977) Dollars Retrievable
Surface facility	19	19
Below-grade facility	7	7
Mine excavation	40	85
Shaft costs	6	6
Backfill	20	40
	92	157
Operating costs/yr	3/yr	3/yr
Decommissioning	5	10

Adding the figures in Table B-26 ($92 million initial capital cost, $3 million/yr operating costs for ten years, and $5 million decommissioning costs) with no discounting yields a cost (1977 dollars) of $127 million, or $860/m^3. An earlier study(51) which assumed a much smaller facility (5000 m^3) developed capital costs of $60 million and annual operating costs of $3.4 million, for unit costs per cubic meter at least 20 times the above figure. Despite this wide range of cost estimates, for every cubic meter of TRU waste volume reduction, there is a potential cost saving of at least $860 for a non-retrievable facility; for a retrievable facility, using figures shown in Table B-26, the cost savings work out to at least $1400/m^3.

REFERENCES

1. Roy, R. et al. Interactions Between Nuclear Waste and Surrounding Rock. Nature 273, May 18, 1978.*

2. Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle ERDA 76-43, U.S. Energy Research and Development Administration, 1976.

3. Dan, G.J. and R.F. Williams.. Status of Commercial Nuclear High-Level Waste Disposal NP-44-SR, 1976.

4. McElroy, J.L. et al. Waste-Solidification Technology U.S.A. In: Proceedings of the International Symposium on the Management of Wastes from the LWR Fuel Cycle. Denver, Colo., July 1976. CONF 76-0701.

5. Techniques for the Solidification of High-Level Wastes, Technical Report, Series No. 176, International Atomic Energy Agency, 1977.

6. Anderson, F.H. et al. Design Criteria for the New Waste Calcining Facility at the Idaho Chemical Processing Plant. In: Management of Radioactive Wastes from the Nuclear Fuel Cycle, Proceeding Series STI/PUB/433, International Atomic Energy Agency, 1977.

7. Freeby, W.A. Fluidized Bed Calcination of Simulated Commercial High Level Radioactive Wastes. In: Radioactive Wastes From the Nuclear Fuel Cycle, AIChE Symposium Series 154:72, 1976.

8. Management of Radioactive Wastes from the Nuclear Fuel Cycle. Proceeding Series STI/PUB/433, International Atomic Energy Agency, 1977.

9. McElroy, J. L., et al. Recent Calcination and Vitrification Process Accomplishments. Proceeding Series STI/PUB/433, International Atomic Energy Agency, 1977.

10. Guber, W., et al. Lab-Scale and Pilot Plant Experiments on the Solidification of High-Level Wastes at the Karlsruhe Nuclear Research Center. Proceeding Series STI/PUB/433, International Atomic Energy Agency, 1977.

11. Gavrenet, A., et al. High Level Waste Management. In: International Conference on World Energy - A Status Report, Proceedings of the Plenary Sessions Volume 25. TANSAO 25 1-257 ISSN: 3-018X, 1977.

12. Barnwell Nuclear Fuel Plant Separations Facility Final Safety Analysis Report. Allied General Nuclear Services, 1974.

13. Blomeke, J.O., et al. Projections of Radioactive Wastes to be Generated by the U.S. Nuclear Power Industry, ORNL TM-3965, Oak Ridge National Laboratory, 1974.

---

*Inserted subsequent to preparation of the draft report

Preceding page blank

14. North, E.D. Solid Waste Generation in Reprocessing Nuclear Fuel. In: Radioactive Wastes from the Nuclear Fuel Cycle, AIChE Symposium Series 154:72, 1976.

15. Moore, R.E. Materials for Water Desalting Plants, Chemical Engineering, October 14, 1963.

16. Daley, G.H. and O.P. Gormley. Handling, Storage, and Disposition of Solid Low-Level Wastes. In: Radioactive Wastes from the Nuclear Fuel Cycle. AIChE Symposium Series 154:72, 1976.

17. Parsons, Brinckerhoff, Quade & Douglas, Inc. Waste Isolation Facility Description Y/OWI/SUB-76/16506, Office of Waste Isolation, 1976.

18. Wolfe, R.A. The Research and Development Program for Transuranic-Contaminated Waste Within the U.S.E.R.D.A. In: Radioactive Wastes From The Nuclear Fuel Cycle. AIChE Symposium Series 154:72, 1976.

19. Blomeke, J.O., and C.W. Kee. Projections of Wastes To Be Generated. In: Proceedings of the International Symposium on the Management of Wastes from the LWR Fuel Cycle. CONF. 76-0701, U.S. Energy Research and Development Administration, 1976.

20. McKenzie, D.E. et al. Mass Reduction of Solid Transuranic Wastes by Molten Salt Combustion. In: Radioactive Wastes From The Nuclear Fuel Cycle. AIChE Symposium Series 154:72, 1976.

21. Lerch, R.E. and J.R. Divine. Waste Volume Reduction by Acid Digestion. In: Radioactive Wastes From The Nuclear Fuel Cycle. AIChE Symposium Series 154:72, 1976.

22. Cooley, C.R. and D.E. Clark. Treatment Technologies for Non-High-Level Wastes (USA). In: Proceedings of the International Symposium on the Management of Wastes from the LWR Fuel Cycle. CONF. 76-0701, U.S. Energy Research and Development Admistration, 1976.

23. Perkins, Betty L. Incineration Facilities for Treatment of Radioactive Wastes: A Review. LA-6252, Los Alamos Scientific Laboratory, July 1978.*

24. Environmental Statement, Transuranium Solid Waste Development Facility, Los Alamos Scientific Laboratory WASH 1537, U.S. Atomic Energy Commission, 1973.

25. Wickland, C.E. Packaging Rocky Flats Waste. Nuclear Technology 32:1, 1977.

26. Moore, J.G., et al. Leach Behavior of Hydrofracture Grout Incorporating Radioactive Wastes. Nuclear Technology 32:1, 1977.

---

*Inserted subsequent to preparation of the draft report

27. Kupfer, M.J., et al. Glass Forms for Alpha Waste Management, In; Radioactive Wastes From The Nuclear Fuel Cycle. AIChE Symposium Series 154:72, 1976.

28. Cheverton, R.D. and W.D. Turner. Thermal Analysis Of The National Radioactive Waste Repository: Progress Through March 1972. ORNL-4789, Oak Ridge, National Laboratory, 1972.

29. Bradshaw, R. and W. McClain. Project Salt Vault: A Demonstration Of The Disposal of High-Activity Solidified Wastes In Underground Salt Mines. ORNL-4555, Oak Ridge National Laboratory, April 1971.

30. Mahtab, M.A. and J.L. Ratigan. Stability of a Radioactive Waste Repository in the Canadian Shield. WS-155-40210, AECL, March 3, 1976.

31. Culler, F.L. Technical Status of the Radioactive Waste Repository. A Demonstration Project for Solid Radioactive Waste Disposal. Testimony Before the Joint Committee. Appendix 17, page 191J.

32. Walsh, E.L. and E.A. Bathke. Thermal Calculations in Shale. OWI/SUB-76/16502, Office of Waste Isolation, September 1976.

33. Parsons, Brinckerhoff, Quade and Douglas, Inc. Thermal Guidelines for a Repository in Bedrock. Office of Waste Isolation, September 1976.

34. Turner, W.D. and M. Simon-Tov. HEATING-3, An IBM 360 Heat Conduction Program. ORNL-TM-3208, Oak Ridge National Laboratory, February 1971.

35. Turner, W.D. and J.S. Crowell. Notes on HEATING--An IBM 360 Heat Conduction Program. CTC-INF-980, Union Carbide Corporation, Nuclear Division, Oak Ridge, Tennessee, November 1969.

36. Levy, S. Use of Explicit Method in Heat Transfer Calculations with an Arbitrary Time Step. Report 68-C-282, General Electric Co., Schenectady, New York, August 1968.

37. Merritt, W.F. High-Level Waste Glass: Field Leach Test. Nuclear Technology 32, January 1977.

38. Mendel, E. High-Level Waste Glass. Nuclear Technology 32, January 1977.

39. Ross, W.A. Properties and Characteristics of High-Level Waste Glass. BNWL-SA-6146.

40. Ames, L.L., D. Rai, and R.J. Serne. A Review of Actinide-Sediment Reactions. BNWL-1983, Battelle Northwest Laboratory, February 10, 1976.

41. Turcotte, R.P. and F.P. Roberts. Phase Behavior and Radiation Effect in High-Level Waste Glass. BNWL-SA-6168, February 1977.

42. Katayama, Y.B. Leaching of Irradiation Fuel Pellets in Deionized and Typical Groundwater. BNWL-2057, 1976.

43. McElroy, J.L. et al. Waste Solidification Program Summary Report, Vol. 11. Evaluation of WSEP High-Level Waste Solidification Processes. BNWL-1667, 1972.

44. Exxon Nuclear Corporation, Nuclear Fuel Recovery and Recycling Center, Preliminary Safety Analysis Report. Docket # 50-564, June 16, 1976.

45. Exxon Nuclear Corporation, PSAR Application Letter, Project #565, February 14, 1976.

46. Cholister, R.J., et al. Nuclear Fuel Cycle Closure Alternatives. Allied-General Nuclear Services. April 1976.

47. Anderson, F.H., et al. Design Criteria for the New Waste Calcining Facility at the Idaho Chemical Processing Plant. In: Management of Radioactive Wastes from the Nuclear Fuel Cycle, Proceeding Series STI/POB/433, International Atomic Energy Agency, 1976.

48. 1000-MWe Central Station Power Plants. WASH-1230, June 1972.

49. Means, R.S., Inc. Building Construction Cost Data, 1977.

50. Radioactive Waste Repository Study. Topical Report 04: Rock Mechanics. Acres Consulting, Ltd., 1976.

51. Raudenbush, M.H. Estimated Costs for Waste Treatment from a 1500 Tonne/Year Reprocessing Plant. S.M. Stoller Corporation Report to Battelle Pacific Northwest Laboratories and Sandia Corporation, October 1976.

GLOSSARY

Actinides: A series of elements in the periodic table, beginning with actinium (element No. 89) and continuing through lawrencium (element No. 103). The series includes uranium (element No. 92) and all of the man-made transuranium elements. All are radioactive.

Alpha particle: A positively charged particle emitted by certain radioactive materials. It is made up of two neutrons and two protons bound together. It is the least penetrating of the three common types of radiation (alpha, beta, gamma) emitted by radioactive material, being stopped by a sheet of paper. It is dangerous to plants, animals, or man only if the alpha-emitting substance has entered the body.

Background radiation: The radiation in man's natural environment, including cosmic rays and radiation from the naturally radioactive elements, both outside and inside the bodies of men and animals. It is also called natural radiation.

Beta radiation: See Decay, radioactive.

Boiling water reactor (BWR): A type of nuclear power reactor that employs ordinary water (H_2O) as coolant and moderator and allows bulk boiling in the core so that steam is generated in the primary reactor vessel.

Breeder reactor: A reactor that produces fissionable fuel as well as consuming it, especially one that creates more than it consumes. The new fissionable material is created by capture (in "fertile" materials) of neutrons from fission. The process by which this occurs is known as breeding.

Calcine: (As a verb) To roast under oxidizing conditions. (As a noun) The solid product of a roasting treatment under oxidizing conditions.

Canister: A metallic container for waste.

Cask: A container for transporting waste canisters or for spent fuel.

Chopper: Device for cutting spent fuel elements into smaller lengths.

Cladding: The outer jacket of nuclear fuel elements. It prevents corrosion of the fuel and the release of fission products into the coolant. Aluminum or its alloys, stainless steel, and zirconium alloys are common cladding materials. Synonym: hull.

Curie: The basic unit to describe the intensity of radioactivity in a material. The curie is equal to 37 billion disintegrations per second, which is approximately the rate of decay of 1 gram of radium. A curie is also a quantity of any nuclide having 1 curie of radioactivity.

GLOSSARY (continued)

Named for Marie and Pierre Curie, who discovered radium in 1898. The prefixes milli-, micro- and nano- are frequently used and indicate quantities of 10^{-3} curie, 10^{-6} curie and 10^{-9} curie, respectively.

Daughter: The nuclide remaining after the radioactive decay of an atomic nucleus. In turn, this daughter may be either stable or radioactive.

Decay heat: The energy released when radioactive nuclides change to their daughter nuclides.

Decay, radioactive: The spontaneous transformation of one nuclide into a different nuclide (or into a different energy state of the same nuclide). The process results in a decrease, with time, of the number of the original radioactive atoms in a sample. It involves the emission from the nucleus of alpha particles, beta particles (electrons) or gamma rays (electromagnetic radiation); or the nuclear capture or ejection of orbital electrons. Also called radioactive disintegration.

Decontamination factor: The ratio of the amount of a given type of radioactive material entering a process (or process step) to that which leaves the process (or process step).

Disposal: Isolating the radioactive waste permanently in a form and manner with no intent to retrieve it.

Element: One of the 103 known chemical substances that cannot be divided into simpler substances by chemical means. A substance whose atoms all have the same atomic number. Examples: hydrogen, lead, uranium. (Not to be confused with fuel element.)

Fertile material: Material in which fissile isotopes can be produced by neutron capture.

Fines: Small-size-range powder.

Fission: The splitting of a heavy nucleus into two approximately equal parts (which are nuclei of lighter elements), accompanied by the release of a relatively large amount of energy and generally one or more neutrons. Fission can occur spontaneously, but usually is caused by nuclear absorption of neutrons or other particles.

Fission products: The nuclei (fission fragments) formed by the fission of heavy elements, plus the nuclides formed by the fission fragments' radioactive decay.

Fuel: Fissionable material used or usable to produce energy in a reactor. Also applied to a mixture, such as natural uranium, in which only part of the atoms are readily fissionable, if the mixture can be made to sustain a chain reaction.

GLOSSARY (continued)

Fuel assembly: A unit containing clad pieces of nuclear fuel for insertion into the core of a nuclear reactor. An integral part of the fuel element is the cladding provided to protect the fuel from corrosion by the reactor coolant and to contain the fission products formed during irradiation.

Fuel bundle: Same as Fuel assembly (q.v.).

Fuel cycle: The series of steps involved in supplying fuel for nuclear power reactors. It includes mining, refining, the original fabrication of fuel elements, their use in a reactor, chemical processing to recover the fissionable material remaining in the spent fuel, re-enrichment of the fuel material, and refabrication into new fuel elements.

Fuel element: Same as Fuel assembly (q.v.).

Fuel reprocessing: The processing of reactor fuel to recover the unused fissionable material.

Fuel rod: A tube containing UO_2 or mixed oxide fuel; part of a fuel assembly.

Gamma radiation: See Decay, radioactive.

Glass ceramic: A ceramic material produced by the controlled devitrification (crystallization) of a glass.

Glassification: Incorporation into glass.

Half-life: The time in which half the atoms of a particular radioactive substance disintegrate to another nuclear form. Measured half-lives vary from millionths of a second to billions of years.

High-level waste: The highly radioactive waste resulting from the reprocessing of spent fuel to separate uranium and plutonium from the fission products. The term includes the high-level liquid wastes (HLLW) produced directly in reprocessing, and the solid high-level wastes (HLW) which can be made therefrom.

Hull: See Cladding.

In-can melting: A process in which solids are converted to a molten glass in the very canister in which it is intended later to bury the glass.

Iodine: An easily-volatilized fission product (q.v.).

Ion: An atom or molecule that has lost or gained one or more electrons. By this ionization it becomes electrically charged. Examples: an alpha particle, which is a helium atom minus two electrons; a proton, which is a hydrogen atom minus its electron.

GLOSSARY (continued)

Isotope: One of two or more atoms with the same atomic number (the same chemical element) but with different atomic weights. Isotopes have very nearly the same chemical properties, but different nuclear (radioactive-decay) properties. Thus, for the element carbon, for example, the isotope of atomic weight 12 (C-12) and the isotope of atomic weight 14 (C-14) behave identically in chemical reactions; but whereas C-12 is not radioactive, C-14 is radioactive, decaying with a 5730-year half-life to stable nitrogen (N-14) with release of a beta particle.

Krypton: A gaseous fission product (q.v.).

Leaching: Extracting material from a solid by contacting it with water or with a solution.

Light water reactor: A reactor in which ordinary water (H_2O) is used as the coolant. In such reactors the water is either allowed to boil (boiling water reactor or BWR) or pressurized to prevent boiling (pressurized water reactor or PWR).

Loading: Amount of waste contained per unit of volume.

Low-level waste: Waste containing types and concentrations of radioactivity such that shielding to prevent personnel exposure is not required.

Matrix: The base material throughout which other materials are dispersed.

Metal matrix: An arrangement whereby aggregates of radioactive matter are dispersed in a continuous metallic block.

Mil: One thousandth of an inch.

Mill: One thousandth of a dollar.

Mixed-oxide fuel cycle: A fuel cycle (q.v.) in which fuel containing both uranium oxide and plutonium oxide is fed to the reactors. Such a cycle requires reprocessing of spent fuel to recover the residual uranium and the plutonium for fabrication of fuel elements.

Monolith: A solid material composed of a single phase, uninterrupted by cracks or voids.

Natural uranium: Uranium as found in nature, containing 0.7% of U-235, 99.3% of U-238, and a trace of U-234. It is also called normal uranium.

Nuclear criticality: The existence of all conditions needed to initiate a nuclear fission chain reaction, as in a nuclear reactor or in an atomic bomb.

Nuclide: A general term applicable to all atomic forms of the elements. The term is often used erroneously as a synonym for "isotope," which properly has a more limited definition. Whereas isotopes are the various forms of a single element (hence are a family of nuclides) and all have the same atomic number and number of protons, nuclides comprise all the isotopic forms of all the elements.

Off-gas: The gas given off in any process step.

Plasma: A gas at a temperature sufficiently high that the gas molecules become completely ionized. This occurs at temperatures of tens of thousands of degrees C or more.

Plenum spring: A small spacing element placed inside the cladding end cap of a fuel rod to maintain the fuel pellets in a stable configuration.

Plutonium: A heavy, radioactive, man-made metallic element with atomic number 94. Its most important isotope is fissionable plutonium-239, produced by neutron irradiation of uranium-238. It is used for reactor fuel and in weapons.

Pressurized water reactor (PWR): A type of power reactor that employs ordinary water (H_2O) as coolant and moderator and is pressurized to keep the exit coolant stream from boiling.

Pyrolysis: Thermal breakdown of a solid or liquid to form gases, in the absence of sufficient air to burn these gases completely.

Rad: A measure, applicable to any form of ionizing radiation, of actual energy absorption, being defined as the amount of energy imparted to matter by ionizing radiation per unit mass of irradiated material at the place of interest. One rad corresponds to the absorption of 100 ergs of energy per gram (100 ergs = 6.24×10^7 million electron volts, Mev). In interpreting standards of radiation protection, it is generally permissible to consider 1-roentgen exposure to gamma or x-rays as roughly equivalent to a dose of 1 rad in soft tissue.

Radiation: The emission and propagation of energy through matter or space by means of electromagnetic disturbances which display both wave-like and particle-like behavior; in this context, the "particles" are known as photons. Also, the energy so propagated. The term has been extended to include streams of fast-moving particles (alpha and beta particles, free neutrons, cosmic radiation, etc.). Nuclear radiation is that emitted from atomic nuclei in various nuclear reactions, including alpha, beta, and gamma radiation and neutrons.

Radioactivity: The spontaneous decay or disintegration of an unstable atomic nucleus, usually accompanied by the emission of ionizing radiation.

Radioisotope: A radioactive isotope. An unstable isotope of an element that decays or disintegrates spontaneously, emitting radiation. More than 1300 natural and artificial radioisotopes have been identified.

Radionuclide: A radioactive nuclide. Thus, carbon-14 (C-14) is a radionuclide because it decays radioactively to nitrogen-14 (N-14).

Radwaste: A contraction of the term "radioactive waste."

Recycle: The returning of uranium and plutonium (recovered in spent fuel reprocessing) for reuse in new reactor fuel elements.

Rem: A dose unit which takes into account the relative biological effectiveness (RBE) of the radiation. The rem ("roentgen equivalent man") is defined as the dose of a particular type of radiation required to produce the same biological effect as one roentgen of (0.25 Mev) gamma radiation. A 1-rad dose of alpha particles is approximately equivalent in its biological effects to 10 rads of gamma radiation, and hence may be expressed as 10 rems. A millirem (mrem) is one thousandth of a rem.

Roentgen (R): A measure of the ability of gamma or X rays to produce ionization in air. One roentgen corresponds to the absorption of about 86 ergs (100 ergs = 6.24×10^7 million electron volts, Mev) of energy from X- or gamma radiation, per gram of air. The corresponding absorption of energy in tissue may be from one-half to two times as great, depending on the energy of the radiation and the chemical composition of the tissue. The roentgen is thus more useful as a measure of the amount of gamma or X rays to which one is exposed than as a measure of the dose of such radiation actually received.

Ruthenium: A sometimes-volatilized fission product (q.v.).

Salt cake: The solid residue resulting from a concentration of high-level liquid waste in underground waste storage tanks.

Salt dome: A geologic salt formation in which a plug of salt has been thrust up through rock at some depth, leading to a subterranean "cylinder" of salt which may be a mile or more in diameter and several miles deep.

GLOSSARY (continued)

Scoping analysis: An analysis made in order to determine the upper and lower bounds of a phenomenon.

Sintering: Agglomeration of particles, achieved by heating them almost to their melting point.

Source terms: The amounts of specific radioactive nuclides issuing from a process or from a process step.

Spalling: Breaking off of small pieces from a larger mass.

Sparge: Bubbling of a gas through a liquid.

Spent fuel: Fuel after its use in a nuclear reactor. It then contains fission products, activation products and actinides, many of which are radioactive. Synonym: irradiated fuel.

Springline: The line of intersection of roof and walls.

Supercalcine: A crystalline ceramic waste form prepared by mixing selected additives with HLLW and then calcining.

Throwaway fuel cycle: One in which the spent fuel is disposed of directly rather than reprocessed.

Transuranic elements: Elements with atomic numbers greater than 92. They include neptunium, plutonium, americium, curium, and others.

Transuranic waste: Any waste material measured or assumed to contain more than a specified concentration of transuranic elements.

Tritium: A radioactive isotope of hydrogen, of atomic weight 3. Tritium (H-3) has a half-life of 12.3 years.

Uplift: The vertical rise on the surface of the earth caused by thermal expansion in the case of a waste repository.

Uranium: A radioactive element with the atomic number 92 and, as found in natural ores, an average atomic weight of approximately 238. The two principal natural isotopes are uranium-235 (0.7% of natural uranium) which is fissionable, and uranium-238 (99.3% of natural uranium) which is fertile. Natural uranium also includes a minute amount of uranium-234. Uranium is the basic raw material of nuclear energy.

Waste, radioactive: Equipment and materials (from nuclear operations) that are radioactive and for which there is no further use.

ABBREVIATIONS

AGNS	Allied Gulf Nuclear Services
ARHCO	Atlantic Richfield Hanford Company
BNFP	British Nuclear Fuels Processing
BNWL	Battelle Northwest Laboratories
BWR	Boiling Water Reactor
CANDU	Canadian Deuterium (Heavy Water) Reactor
DF	Decontamination Factor
EIS	Environmental Impact Statement
ERDA	Energy Research and Development Administration
EPA	Environmental Protection Agency
FP	Fission Product
GE	General Electric
HEDL	Hanford Engineering Development Laboratory
HLLW	High-Level Liquid Waste
HLW	High-Level Waste
HM	Heavy Metal (Uranium and Plutonium)
HTGR	High Temperature Gas-Cooled Reactor
HWR	Heavy Water Reactor
ICPP	Idaho Chemical Processing Plant
ILLW	Intermediate-Level Liquid Waste
INEL	Idaho National Engineering Laboratory
LASL	Los Alamos Scientific Laboratory
LMFBR	Liquid Metal Fast Breeder Reactor
LTA	Low Temperature Adsorption
LWR	Light Water Reactor
MTU	Metric Ton of Uranium
NRC	Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
ORGDP	Oak Ridge Gaseous Diffusion Plant
PVC	Polyvinyl Chloride
PWR	Pressurized Water Reactor
TRU	Transuranic
WSEP	Waste Solidification Engineering Project

Å	Angstrom unit (10^{-8} cm)
Ci	Curie
d/m	Disintegrations per minute
GW	Gigawatts (1 GW = 10^6 kW)
GW-yr	Gigawatt-year
kg	kilogram
kW	kilowatt
m	Meter
mCi	Millicurie
MCi	Megacurie
µCi	Microcurie
mR	milliRoentgen
mrem	millirem

ABBREVIATIONS (continued)

MT	Metric ton
MTHM	Metric ton of heavy metal (Uranium and Plutonium)
MW_e	Megawatts electrical
MW_t	Megawatts thermal
MW_t-yr	Megawatt thermal-year
nCi	Nanocurie

APPENDIX B-II

SOLIDIFICATION PROCESSES IN EARLY STAGES OF DEVELOPMENT

B-II.1 SUPERCALCINE

ERDA 76-43 describes supercalcine as a crystalline ceramic waste form prepared by mixing selected additives with high-level liquid waste (HLLW) and then calcining(1). The result is a mixture of thermodynamically stable crystalline phases that are more refractory and leach-resistant than ordinary oxide calcine formed without additives.

Work on the development of supercalcine is being conducted at Pennsylvania State University, fixing HLLW in stable inert crystalline phases. Waste streams containing 35-45% by weight of additives, usually including Ca, Sr, Al, and Si, can be calcined by any of the previously described methods. One nonradioactive engineering-scale run has been made at BNWL. Figure B-II-1 shows two of a number of possible options for processing waste as a supercalcine.

Several more years will be required to develop the full potential of the supercalcine process for all HLLW constituents to be incorporated into optimally stable crystalline forms.

If the supercalcine is part of a composite waste form, several integrated processing steps, such as pelletizing and application of coating, must be developed. It may be ten years before a practical design can be developed that would use supercalcine for the core of a multi-barrier waste form.

Laboratory work is continuing in the following areas:

- Refinement of additives and formulations.
- Reduction of potential volatilization losses of radioactive materials not currently included in crystalline phases.
- Development of consolidation methods for supercalcine.
- Studies of leachability and thermal stability.
- Assessment of radiation effects.

B-II.2 SINTERING PROCESSES

If calcine is mixed with the proper flux or frit and sintered, the product will have both a glass and crystalline phase and will be more leach resistant, have high impact strength, be resistant to thermal

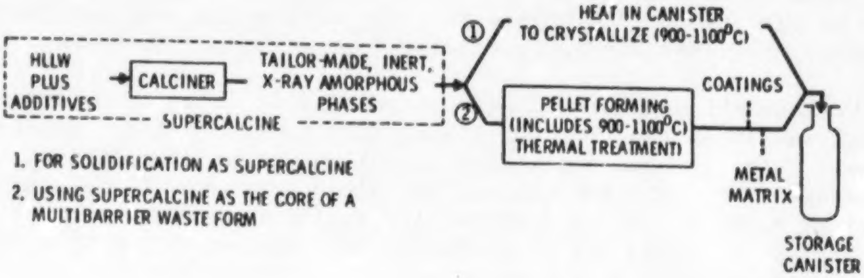

HLLW
PLUS
ADDITIVES
CALCINER
TAILOR-MADE, INERT,
X-RAY AMORPHOUS
PHASES
SUPERCALCINE
①
HEAT IN CANISTER
TO CRYSTALLIZE (900-1100°C)
②
PELLET FORMING
(INCLUDES 900-1100°C)
THERMAL TREATMENT)
COATINGS
METAL
MATRIX
STORAGE
CANISTER
1. FOR SOLIDIFICATION AS SUPERCALCINE
2. USING SUPERCALCINE AS THE CORE OF A MULTIBARRIER WASTE FORM

FIGURE B–II–1 SUPERCALCINE PROCESSES

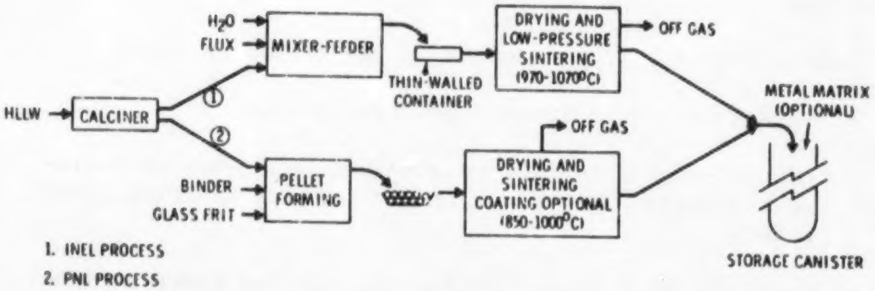

H_2O
FLUX
MIXER-FEEDER
THIN-WALLED
CONTAINER
DRYING AND
LOW-PRESSURE
SINTERING
(970-1070°C)
OFF GAS
HLLW
CALCINER
①
②
BINDER
GLASS FRIT
PELLET
FORMING
DRYING AND
SINTERING
COATING OPTIONAL
(850-1000°C)
OFF GAS
METAL MATRIX
(OPTIONAL)
STORAGE CANISTER
1. INEL PROCESS
2. PNL PROCESS

FIGURE B–II–2 SINTERING PROCESSES

Source: Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle. ERDA 76-43, U.S. Energy Research and Development Administration, May 1976.

shock, and remain solid at temperatures up to 800°C. The product can be formed as pellets or as large thick discs.

The Idaho National Engineering Laboratory (INEL) and BNWL processes for producing these sintered glass ceramics (SGC) are illustrated in Figure B-II-2.

Frits used by INEL and BNWL differ somewhat(1). Product densities range from 2.4-3.3 g/cm^3. Specific volumes of the reference waste would be 30-55 liters/MTU. The leach rates of well-sintered products are similar to those of glass.

Various methods of preparing the material for sintering and various post-treatments are under investigation. Work thus far has been on a laboratory scale only; future work will concentrate on development of process equipment and design verification. Experimental development may be required for product forming, moving the containers through sintering furnaces, and off-gas cleanup. Pilot-plant operation is scheduled for 1977. More work must also be done on product characterization. The design of a full-scale demonstration plant may be started in 1979 or 1980.

B-II.3 METAL MATRICES

Metal matrices are monoliths with high thermal conductivity composed of calcine, vitreous beads, or pellets cast in molten metal or embedded in sintered metal. Alloys of lead, zinc, aluminum, or other low-melting metals are used in casting; iron, iron alloys, or copper are used in sintering. Waste constitutes up to 65% of the matrix.

The main advantage of these matrices is that they provide a high-conductivity form to decrease centerline temperatures. This would tend to make them advantageous for use with "young" wastes where the heat dissipation problems are more severe. Their stability may be limited to temperatures below the melting point of the metal to minimize slow oxidation or reaction of the metal with the waste or canister. The matrices vary in leach resistance, depending on the form of the waste incorporated.

Figure B-II-3 is a schematic illustration of some methods of forming a metal matrix.

Cast matrices can be formed either by introducing the formed waste near the bottom of the canister and letting it displace the molten metal, or by forcing molten metal by pressure or vacuum through a canister of waste particles. Sintered matrices can be formed either by mixing wastes and metal powder, compacting the mixture into a pellet and sintering, or by vibrating a metal powder throughout a canister loaded with formed waste and sintering the mixture into a coherent mass.

Matrices have been formed and tested on a laboratory scale. Casting of vitreous beads by molten metal displacement has been verified on a nonradioactive pilot-plant scale.

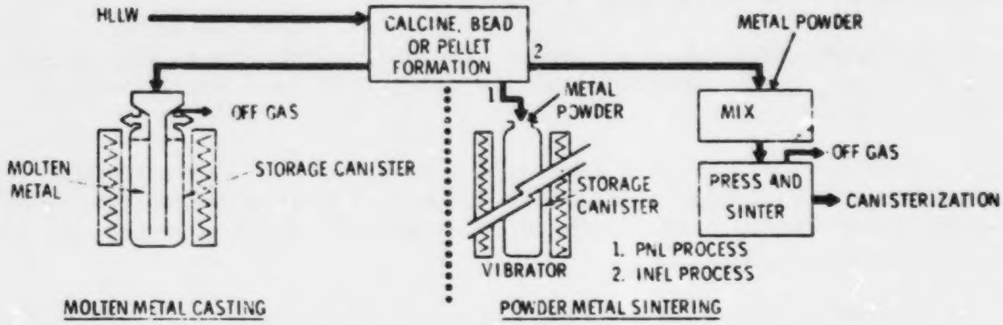

HLLW
CALCINE, BEAD OR PELLET FORMATION
METAL POWDER
2
1
METAL POWDER
MIX
OFF GAS
OFF GAS
MOLTEN METAL
STORAGE CANISTER
STORAGE CANISTER
PRESS AND SINTER
CANISTERIZATION
1. PNL PROCESS
2. INEL PROCESS
VIBRATOR
MOLTEN METAL CASTING
POWDER METAL SINTERING

FIGURE B–II–3 METAL MATRIX FORMATION

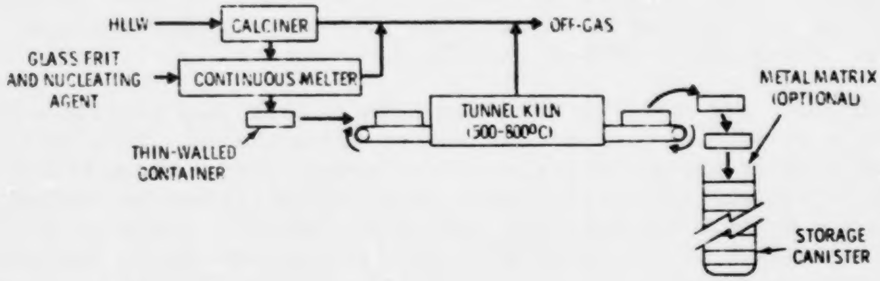

HLLW
CALCINER
OFF-GAS
GLASS FRIT AND NUCLEATING AGENT
CONTINUOUS MELTER
THIN-WALLED CONTAINER
TUNNEL KILN (500-800°C)
METAL MATRIX (OPTIONAL)
STORAGE CANISTER

FIGURE B–II–4 GLASS CERAMIC PROCESS

Source: Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle. ERDA 76-43, U.S. Energy Research and Development Administration, May 1976.

ERDA 76-43[1] lists the major development efforts required:

- Evaluation of matrix formation properties of new waste compositions.
- Development of best methods for use under remote operating conditions.
- Investigation of methods for casting fines or reducing percentage of fines.
- Verification of methods of metering particles and molten metal to the casting container.
- Determination of best process techniques for receiving waste, canister hookup, and removal from matrix-forming apparatus.
- Study of methods for compacting mixes for sintering operations.
- Continuation of product characterization studies.
- Operation of non-radioactive pilot plant to verify a conceptual process.

Design for a full-scale facility could start in late 1978 or early 1979.

B-II.4 GLASS-CERAMICS

If a glass is subjected to a controlled crystallization, a fine-grained crystalline body with some residual glass phase may be produced. This glass-ceramic is stronger than glass, is not as susceptible to uncontrolled devitrification at high storage temperatures, and has leach resistance similar to that of glass. A thin cross section (3 cm or less) is needed to allow rapid cooling and to maintain uniform temperatures during processing, in order to control nucleation and crystal growth. One acceptable geometry would be a disc, which could be stacked into a canister with a filler (probably metal), to aid heat transfer between the discs and canister walls.

Because work thus far has been limited to laboratory scale development, the process is only a concept at present. It is illustrated in Figure B-II-4.

To develop the best compositions and characterize their behavior during the waste fixation and storage process would take 2-5 years of major laboratory research. Engineering development of the glass-casting and heat treating processes would also require a major effort(1).

Because there is now no such major effort in the United States to develop this process, although work has been done in Germany, it is difficult to estimate when a production facility could be designed. It would probably be well after 1980.

B-II.5 COATED PELLETS

Leach resistance of solidified wastes can be increased by forming the waste into pellets and coating the pellets with a variety of materials, including pyrolytic carbon, SiC, SiO_2, and Al_2O_3. The coatings could be applied by chemical vapor deposition in a fluidized-bed or drum coater, by plasma spray, or by an enameling process.

Because the coated pellets have poor thermal conductivity and their coatings could chip during handling and transport, it may be desirable to incorporate the pellets into a metal matrix.

The possible combinations of waste form, coating type, and composition of metal matrix are still being evaluated. Besides the process problems that must be overcome in determining which of the above choices is best, the additional handling involved in moving materials from calciner to pellet-former to coater to canister will create more complications. The resultant complexity may affect process reliability.

ERDA 76-43(1) identifies the following major developmental efforts required:

- Selection of pelletizing and coating process,
- Optimization of the matrix and characterization of the product, and
- Design of equipment for remote operation.

It will probably be five years or more before conceptual design can begin.

B-II.6 ION EXCHANGE

Sandia Laboratories is developing a method of disposing of high-level radionuclides by first fixing them on ion-exchange media and then compacting and sintering the media. A simplified flowsheet of the process is shown in Figure B-II-5.

The HLLW is neutralized by the addition of NaOH and clarified by centrifugation. The clarified waste is then almost completely decontaminated by ion exchange with sodium titanate (ST). On a laboratory scale, the effluent from the ST ion exchange has been reduced to less than 10^{-5} Ci/ml of total activity; most of the carryover radioactivity

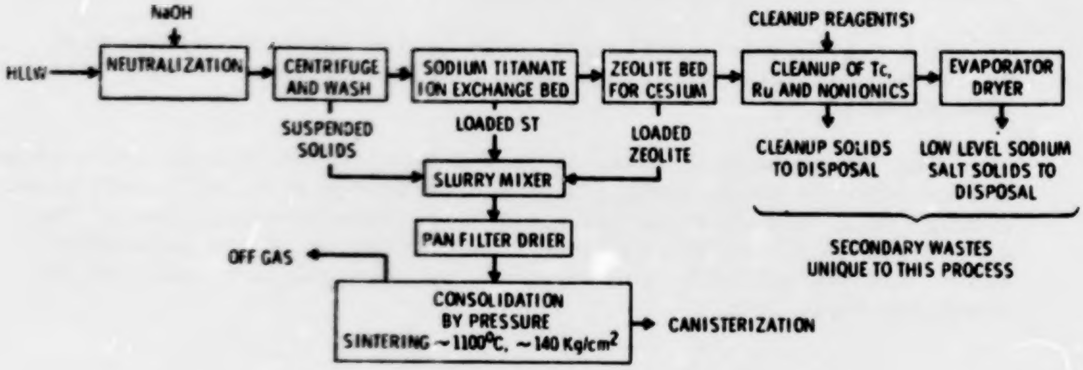

NaOH
HLLW
NEUTRALIZATION
CENTRIFUGE AND WASH
SODIUM TITANATE ION EXCHANGE BED
ZEOLITE BED FOR CESIUM
CLEANUP REAGENT(S)
CLEANUP OF Tc, Ru AND NONIONICS
EVAPORATOR DRYER
SUSPENDED SOLIDS
LOADED ST
LOADED ZEOLITE
CLEANUP SOLIDS TO DISPOSAL
LOW LEVEL SODIUM SALT SOLIDS TO DISPOSAL
SLURRY MIXER
SECONDARY WASTES UNIQUE TO THIS PROCESS
PAN FILTER DRIER
OFF GAS
CONSOLIDATION BY PRESSURE SINTERING ~1100°C, ~140 Kg/cm²
CANISTERIZATION

FIGURE B–II–5 ION EXCHANGE FIXATION

Source: **Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle. ERDA 76-43, U.S. Energy Research and Development Administration, May 1976.**

is Ru, Cs, and Tc. Activity of alpha emitters was reduced to less than 10^{-10} Ci/ml. Cesium may be subsequently removed by ion exchange with a zeolite, and remaining traces of such wastes may be removed with an anion exchanger material and other reagents. The spent ST and zeolite are removed as slurries, combined with the suspended solids removed in the clarification step, dewatered and dried on a pan filter, and then consolidated into cylinders by pressure sintering.

The major process effluents would be low-level dry sodium salt solids and tritiated water. A small quantity of trace cleanup solids is also expected.

The ion exchange process is in the bench-scale demonstration stage and is expected to be ready for practical design of a full-scale plant during 1981. The experimental program includes investigation of the effectiveness of the ion exchanger, consolidation of the solidified waste, and evaluation of the physical and chemical properties of the resulting waste form.

B-II.7 GERMAN THERMITE PROCESS

A thermite process is being developed in Karlsruhe as a possible alternative to the glass melting step in the German VERA process.

The thermite reaction produces a rugged, two-phase, ceramic-metal material, with properties at least equivalent to glass.

A 2-kg batch process has been tested, and an apparatus has been built for processing batches up to 20 kg to test the process on a larger scale.

B-II.8 GERMAN BOROSILICATE GLASS PROCESS

Work at Julich is directed toward perfecting a denitration, drum-drier, in-can melting process for borosilicate glass.

Formaldehyde is added to the liquid waste until the pH rises to about 3.0; then glass-forming additives ($CaCO_3$, SiO_2) are introduced, which form a viscous slurry. The slurry is converted to a solid cake that is continually scraped off the drier and dropped into a canister where it is melted. This process has been demonstrated on a laboratory scale for both radioactive and nonradioactive glasses.

B-II.9 GERMAN PHOSPHATE GLASS PROCESS

The Pamela process is being developed by Gelsenberg AG of Germany in cooperation with the Eurochemic Company of Belgium. It is being supported by the German Ministry of Research and Technology.

HLLW is denitrated with formaldehyde and concentrated with H_3PO_4 to form a phosphate glass. Originally a platinum melter was used; a later version uses an electrically-heated ceramic melter. The glass melt passes through a series of nozzles and the droplets are caught on a rotating disc, where they solidify. The granulated glass is encapsulated in a lead alloy using technology developed at Eurochemic. Vitrification off-gas is fed back into the reducing atmosphere of the denitration unit, which retains any ruthenium that has volatilized.

Granules of phosphate glass that may contain about 30% by weight of fission product oxides have been favored so far. Work is under way, however, to extend the process to the production of borosilicate glass and borosilicate glass-ceramic products.

B-II.10 OTHER FOREIGN PROCESSES

Several solidification processes are currently being developed in Russia, Italy, Japan, and Sweden. They combine features of the processes already discussed. They will therefore not be described in this report.

REFERENCE

1. Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle. ERDA 76-43, U. S. Energy Research and Development Administration, May 1976.

APPENDIX B-III

OFF-GAS TREATMENT TECHNOLOGY

This appendix discusses in detail the technology available for various degrees of control of five gaseous materials generated during solidification processes (see Section B-3.2.4 of the text):

1. Krypton, with its Kr-85 content;
2. Iodine and its compounds, with their I-129 content;
3. Carbon compounds, with their C-14 content;
4. Hydrogen and water vapor, with their tritium (H-3) content; and
5. Ruthenium compounds, with their Ru-106 content.

Each of these is presented in a separate section of this Appendix.

B-III.1 KRYPTON

ERDA 76-43 gives annual Kr-85 production from a 1500-MT/yr LWR reprocessing plant as 9.8 x 10^6 Ci, or about 2 x 10^5 Ci/GW-yr.(1) These figures (equivalent to about 6500 Ci/MT of fuel) are somewhat lower than another estimate of 9700 Ci/MT.(2) Because Kr-85 has a moderately long half-life (10.7 years), and is produced in large quantities, the U. S. Environmental Protection Agency (EPA) has promulgated for the entire fuel cycle Kr-85 discharge limits of 50,000 Ci/GW-yr.(3) Although the need for these limits has been debated, since krypton is chemically inert and does not present an internal biological hazard, such considerations are beyond the scope of this study.

When the EPA-proposed regulation is used and the less than 1% of krypton that escapes at the reactor plant is ignored, the reprocessing plant decontamination factor (DF) required to meet the 50,000 Ci/GW-yr limit is: $2 \times 10^5/(0.5 \times 10^5) = 4$.

Because krypton is a noble gas, it is reasonable to assume that essentially all the krypton is given off as "off-gas" at the chopper and dissolver steps and thus, is readily removed into the off-gas stream.

According to ERDA 76-43, krypton will be present in the off-gas in concentrations of about 0.003% by volume; xenon will be present in concentrations of 0.02% by volume.(1) Thus, technology for krypton removal must be capable of removing krypton present in the off-gas in small quantities; the required decontamination factor to meet EPA standards is modest (about 4).

Two technologies for krypton removal have been investigated: cryogenic distillation, and fluorocarbon absorption. Two technologies for krypton storage have been investigated: pressurized cylinder storage and zeolite encapsulation.

B-III.1.1 Cryogenic Distillation

This process, identified in ERDA 76-43 as "currently available" technology, is the most commercially advanced of the two krypton removal methods.(1) Cryogenics are used extensively for commercial gas separation. At the Idaho Chemical Processing Plant (ICPP), cryogenic distillation has been used to obtain krypton from off-gas, though not at high overall system efficiencies.

The boiling point of krypton (-157°C) is sufficiently higher than that of oxygen (-183°C), nitrogen (-196°C), and carbon monoxide (-190°C) to allow separation from these gases. However, gases will probably be present with a boiling point higher than -157°C (N_2O, -90°C; NO_2, 21°C; CO_2, -79°C; H_2O, 100°C) and may present freezing problems. Xenon, with a boiling point of -107°C, and NO, with a boiling point of -152°C, will tend to follow the krypton through the cryogenic process but can be separated if desired.

At the cryogenic plant at Idaho, a catalytic converter (rhodium, 480°C) converted N_2O to $N_2 + O_2$, and a cooler/drier (silica gel) removed residual water and NO_2. Nonetheless, considerable difficulty was reported with plugging of the cold trap and other process equipment due to CO_2, H_2O and NO_x freezing, a difficulty that could probably be overcome or the effects mitigated in a commercial-scale plant with increased regeneration/redundancy design and surge volume.

The Idaho plant (a customized, uniquely designed unit) utilized a continuous primary distillation column and periodic transfer of bottoms (containing primarily noble gases with NO if present and some air carryover) to a batch distillation column for final concentration of the krypton. The equipment at the ICPP facility distilled krypton at an efficiency of 97% and xenon at an efficiency of 98%. However, the whole system efficiencies during operating periods were 52% and 63%, respectively. These lower numbers should be viewed with the understanding that the ICPP facility was designed to allow some krypton removal for commercial use, not for environmental protection. Thus, high overall operating efficiency was not a design objective: "The discrepancy between overall system efficiency and cryogenic equipment efficiency is due mainly to losses occurring during unanticipated process interruptions and during startup-shutdown periods."(4) Nonetheless, the efficiencies achieved indicate that a distillation DF of ten is well within reason for a commercial facility, although there is considerably less certainty regarding the ability of a commercial facility to operate with sufficiently high reliability to yield a system DF greater than ten. The Idaho facility treated 240 liters/min (8.4 standard ft^3/min) of dissolver off-gas at atmospheric pressures and 60°C.(4)

The product of the ICPP krypton facility was typically high in contaminants (the product was shipped to Oak Ridge National Laboratory [ORNL] for final purification). Of five runs reported, the average content was: 23% Kr, 60% Xe, 4% N_2, 10% O_2, 2% Ar, and detectable CO_2 and NO_x.(4) The highest krypton content was 51%, but this run contained 25% O_2, a potentially undesirable storage situation.

Considerable concern has been expressed regarding the potential dangers of ozone production in a cryogenic facility. The main point of concentration of the ozone appears to be in the primary distillation column, where liquid oxygen can accumulate and be converted to ozone via radiolysis (from the radioactive krypton). At cryogenic temperatures, the ozone will be decomposed less easily. Sources at the Idaho laboratory suggested that the ozone problem in storage bottles is less significant because of the higher rate of decomposition (confirmed by tests) at or above ambient temperatures.

There are two options for dealing with the ozone problem in a cryogenic system. The oxygen may be removed ahead of the distillation process, or the process may be designed so that residence times of liquid oxygen with radiokrypton are minimized. At ICPP, the latter was done; it is not clear that design of a commercial-scale facility would permit this. Removal of oxygen and ozone has been performed with an O_2/H_2 recombiner; potential difficulties exist with this concept, including the H_2 explosion potential, and greater potential for freeze-plugging of the system by reduction in the O_2 level (NO_x and noble gases are less soluble in LN_2 than in liquid oxygen).

B-III.1.2 Fluorocarbon Absorption

Somewhat less commercially developed, but with fewer apparent problems, is the fluorocarbon system, whereby a fluorocarbon ("R-12": dichlorodifluoromethane) selectively absorbs impurities, which are then stripped from the fluorocarbon.

At the Oak Ridge Gaseous Diffusion Plant (ORGDP), pilot plant tests at 210-620 liters/min (7.5-22 standard ft^3/min) have been performed with a fluorocarbon system that uses radioactive krypton in nitrogen feed gas containing various amounts of O_2, CO_2, nitrogen oxides, iodine, and xenon.(5) The pilot plant consisted of a primary absorption/stripping operation and peripheral equipment required for recovery, purification, and recycle of solvent and some purification of krypton product.

The advantage of the fluorocarbon system is that Kr, CO_2, NO_2, CH_3I, and I_2 all have substantially higher affinity for R-12 than the main off-gas components O_2 and N_2. Xenon also will be selectively absorbed by the R-12. This characteristic makes the system potentially very attractive as an integrated Kr-85, C-14, I-129 removal system.

The tests at the ORGDP pilot facility yielded krypton DFs of greater than 10^3, CO_2 removal DF's of greater than 10^4, iodine DFs (both CH_3I and I_2) of greater than 10^4, and NO_2 removal of greater than 100. The stripper product gas will not be pure Kr but will contain quantities (depending on feed gas) of Xe, Ar, and CO_2. The iodine, NO_2, and H_2O may be removed in the solvent purification step. Figure B-III-1 shows the various gas exhaust points. A program is in progress to determine ways of removing contaminants from the stripper off-gas. ORGDP representatives state that argon may be desirable as a diluting gas for krypton storage, though this is not clear since only 6% of the krypton is expected to be Kr-85.(1)

Some O_2, N_2, and R-12 vapor will also be present in the product gas. A cold trap could be used to trap the CO_2, Xe, and R-12, though not the O_2, N_2, or Ar. The CO_2 could then be removed from the cold-trap regenerate in a caustic scrubber (see section on carbon-14). Alternatively, an inline CO_2 caustic scrubber could be used. A reactive metal trap (copper or manganese) may be tested for O_2 removal, and a titanium metal trap for N_2 removal. The product purification features have not been integrally tested at the pilot plant.

Iodine, NO_2, and H_2O will remain in the R-12 through the stripper and are removed in the solvent still. I_2, CH_3I, NO_2, and H_2O leave the solvent still as liquids or solids in solution, and several tests at ORGDP have confirmed the removal of iodine at high DF's. Separation of the iodine from NO_2 and H_2O has not been developed at ORGDP, and it is not clear that this is a simple step. Use of a solid adsorbent has been suggested, but this technique has economic drawbacks for total iodine removal (see B-III.2). Cold-freezing has been suggested, but it does not appear this would be successful for CH_3I. An alternative solution may be removal of iodine prior to the fluorocarbon process.

The ORGDP pilot plant has accumulated over 6000 hours of operating time. A reliability analysis has shown that equipment availability is 90% at the pilot facility; a program is currently underway to determine what design features are necessary for a 99% availability at a demonstration facility.

Automatic control response to variations in feed gas flows and composition has not been tested completely.

B-III.1.3 Low-Temperature Adsorption

Although this method is not discussed in ERDA 76-43, low-temperature adsorption (LTA) is worth mentioning in any discussion of noble gas removal. At ICPP an LTA process for noble gas removal was investigated but abandoned because of questions of efficiency and freeze-up problems with contaminants such as H_2O, CO_2, NO_2, and N_2O. General Atomic Company has successfully used an LTA system for cleanup of helium streams in a high-temperature gas-cooled reactor (HTGR) application, but H_2O

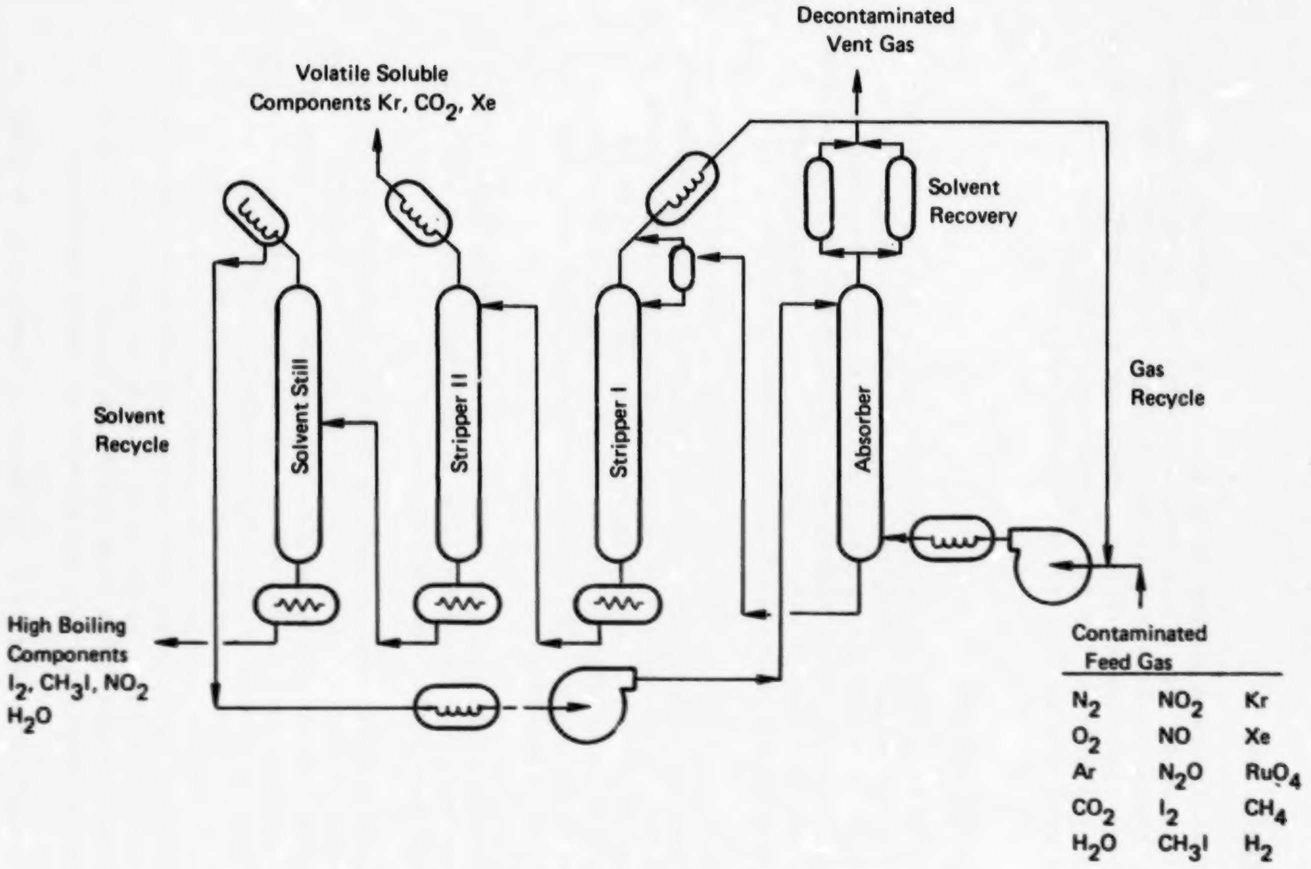

Decontaminated Vent Gas
Volatile Soluble Components Kr, CO_2, Xe
Solvent Recovery
Gas Recycle
Solvent Recycle
Solvent Still
Stripper II
Stripper I
Absorber
High Boiling Components I_2, CH_3I, NO_2 H_2O
Contaminated Feed Gas
N_2 NO_2 Kr
O_2 NO Xe
Ar N_2O RuO_4
CO_2 I_2 CH_4
H_2O CH_3I H_2

FIGURE B–III–1 SCHEMATIC DIAGRAM OF THE ORGDP SELECTIVE ABSORPTION PILOT PLANT

Source: Stephenson, M.J. and R. J. Eby. Development of the 'FASTER' Process for Removing Krypton-85, Carbon-14, and Other Contaminants. Presented at the 14th ERDA Air Cleaning Conference, August 1976.

at a concentration of more than a few parts per million rapidly plugs the cold-traps.

B-III.1.4 Other Systems

ORNL has investigated a number of krypton-removal systems and concluded: "Based on the preceding review of krypton recovery methods, it appears that there are only two processes which are suitable for adoption to a fuel reprocessing plant. These are fluorocarbon absorption and cryogenic distillation. The other processes either suffer from major technical problems or lack of development effort."(6)

B-III.1.5 Cylinder Storage

Because of the long history of commercial use of pressurized cylinders for storage of gases, including highly corrosive gases, the technology for storage of krypton in pressurized bottles may be considered to be available. However, some additional testing needs to be completed on the specific application of steel cylinders to intermediate-term (100-year) storage of the specific krypton removal product.

Steel-cylinder storage has the advantages of commercial experience (including storage of toxic gases, with use of safety techniques such as packless valves, valve caps, and ultra-conservative pressure margins with non-venting cylinders), long life (at least 500 years has been estimated from testing of 1000 cylinders), good thermal and radiation resistance, low cost, and ease of recovery. Use of stainless steel would add corrosion resistance.

The main disadvantage, the potential for sudden catastrophic failure, is not a serious one since total failure of any one cylinder would not result in significant public health hazard.

The remaining uncertainties regarding cylinder storage mainly involve the internal corrosion effects of non-krypton contaminants in the stored gas: ozone (if present), oxygen, NO_x, and water. An optimum cylinder curie-loading must be determined on the basis of allowable heat rate and resultant corrosion rates. The cryogenic distillation process used at Idaho, for example, leaves substantial O_2 in the product gas. The contaminant corrosion problem would be minimized by additional product purification, or by a probably more cost-effective approach, monitoring of sample cylinder performance during storage life. Any resultant problems (unexpectedly high corrosion rate) could be dealt with by transfer to new cylinders, a relatively simple operation.

Mention has also been made of solid daughter (rubidium) buildup on the cylinders as a disposal problem.(7) For example, a 500-psi cylinder with an initial loading of 128,000 Ci Kr-85 will, after 100 years, contain 0.3 kg of rubidium (as a solid at ambient temperature).

Rubidium has chemical characteristics similar to those of sodium, potassium, and cesium: it will react vigorously with water and will ignite spontaneously in air. Studies of sodium storage in LMFBRs suggest that rubidium will not chemically attack the cylinders.

A cylinder test program is currently in progress at Idaho, with test specimens subjected to various conditions of pressure, radiation (gamma), and temperature.

B-III.1.6 Zeolite

Zeolites are crystalline aluminosilicates with uniform arrays of pores; some are available with 4 Å pore size. A krypton atom at ambient temperature has a diameter of about 3.5 Å. Laboratory tests with pure krypton have shown that at elevated temperatures and pressures, krypton will diffuse into the pores; at ambient temperatures, the krypton will remain in the pore structure.(8) About 1.6 times more storage volume would be required with zeolite than with 500-psi cylinders.(9) The rate of release from the cylinder in an accident would, of course, be less if zeolite encapsulation were employed.

The atoms of other gases have diameters close to or less than krypton (Ar: 2.9 Å; CO_2: 3.3 Å; N_2: 3.2 Å; O_2: 3 Å), and, therefore, presumably would also be trapped in the zeolite. Xenon is slightly larger (3.7 A) and could conceivably be discriminated against. There is some question as to whether in a high-xenon environment, the xenon might not interfere with krypton capture. The effect of rubidium buildup is also not clearly defined, although sodium experiments suggest that rubidium would not be a problem.

B-III.1.7 Other Storage Systems

Clathrates have been investigated as krypton-stabilizing substances.(9) Water solubility, susceptibility to oxidation, and thermal instability make clathrates appear to be unpromising.

B-III.1.8 Summary: Krypton Recovery and Storage

Despite the successful operation of a cryogenic distillation krypton removal facility at ICPP, there are a number of potential difficulties in applying the technology to a large commercial facility in order to meet the 50,000 Ci/GW-yr limit. The main question is expected reliability and derives from the freeze-up problems experienced at ICPP. Provision of redundant trains and rapid regeneration equipment, or large gas surge capabilities, or front-end treatment equipment for H_2O, CO_2, and NO_x, could probably increase overall system reliability.

On a test scale, the fluorocarbon system appears to be more promising than cryogenic distillation and has the added potential advantage of economical removal of CO_2 and perhaps iodine. However, the technology of fluorocarbon absorption is not yet fully developed, even at a test scale, especially at the product purification steps. Nevertheless, reliability analyses have been encouraging regarding the potential system availability in commercial application.

Because cylinder storage is an existing, proven technology, with ease of transfer of stored gas in the event of unexpected difficulties, this appears to be the most reasonable technology to assume for krypton storage for the time frame required for an isotope with short (10.7-yr) half-life.

B-III.2 IODINE

After nuclear fuel has cooled for several months, the only radioisotope of iodine from a fuel reprocessing plant that is of concern is I-129 (1.6×10^7 yr half-life). Estimated uncontrolled release of I-129 from a 5 MT/day LWR fuel reprocessing plant is 3.8×10^5 g/yr, which is more than the estimated pre-1940 worldwide inventory of 2×10^5 g.(10) This estimated uncontrolled release of I-129 from a 5 MT/day plant translates to 60 Ci/yr, or about 1.2 Ci/GW-yr and is consistent with the estimate given in Reference 2a.

The EPA has promulgated regulations limiting discharge of I-129 from the entire fuel cycle to 5 mCi/GW-yr.(3) Calculations show that less than 10^{-6} Ci/yr of I-129 is released from a typical modern LWR.* Thus, it is reasonable to assume that the only significant release of I-129 is from the reprocessing plant. The proposed regulation requiring removal of all but 5 mCi/GW-yr means that only 5 mCi/1.2 Ci or 0.4% of the I-129 may be released from the fuel reprocessing plant. This decontamination factor of 250 may present a difficult engineering problem.

Estimates have been made that over 99% of the iodine can be volatilized into the process off-gas (at the dissolver step).(11) This estimate is based on laboratory and hot cell tests at ORNL with spent LWR fuel and use of an air sparge on the dissolver, and establishment of optimum temperatures, acidity, and residence time.** It cannot be assumed that production-scale processes can achieve greater than 99% iodine removal at the dissolver, and even if 99% were removed and collected, over one-half of the remaining 1% of the iodine would have to be accounted for in order to meet the proposed regulations.

---

*If 0.1% failed fuel is assumed, 1.2 mCi/GW-yr of I-129 will leave the fuel at an LWR, most of which will be trapped in managed waste streams.

**On the basis of hot cell tests, it has been estimated that more than 99% of the radioiodine in the dissolver solutions could be removed,(12) but it is questionable whether large-scale commercial facilities could achieve this level of dissolver removal. Savannah River and ORNL have achieved 99.5% in LWR fuel tests, but these tests used once-through nitric acid, with no recycle system.

Because of the volatility of iodine, more than one-half the remaining 1% may enter the off-gas stream through evaporator/concentrator overheads downstream of the dissolver. Meeting EPA requirements may require treatment of these overhead streams for iodine removal. Because iodine removal efficiencies required to meet these requirements are much higher than has ever been required before, there is little information defining where the iodine will migrate after the dissolver, and this may pose problems. Regardless of how the above potential regulatory problems are resolved, very high iodine DFs are clearly desirable in the reprocessing off-gas system.

In addition to the approximately 380 kg of I-129 produced yearly from a 5 MT/day plant, approximately 250 kg of stable I-127 from fission and from reprocessing chemicals will also be present in the off-gas stream.(11) Thus, a design objective in potential iodine control systems is the removal of approximately 600-650 kg of iodine per year at very high decontamination factors (10^3 or better). Approximately 1-5% of organic iodine (methyl iodide, CH_3I) may be present in the off-gas stream, as well as HI and HOI.(2a.)

Four possible technologies have been identified for iodine removal: caustic scrubbing, mercuric nitrate-nitric acid scrubbing (Mercurex), nitric acid scrub (Iodox), and chemisorption by metal-loaded adsorbents.(1)

B-III.2.1 Caustic Scrub

Caustic scrubbing has been used at both Idaho and Hanford for iodine removal, but this method has been generally abandoned in planning for iodine removal from commercial reprocessing plants, because of the very low efficiency of removal of organic iodine forms (which may constitute up to 5% of the iodine) and the low DF for elemental iodine (about 10).(1) In addition, high volumes of waste are generated (a 60 m^3/min gas-flow scrubber would produce 1800 m^3/yr of waste solution(1)). There are no active programs in the United States to develop caustic scrub for iodine removal from reprocessing off-gas streams.

B-III.2.2 Mercurex*

The Mercurex process utilizes a mercuric nitrate-nitric acid scrub in a packed tower, with periodic transfer of the scrub solution to a concentrator-evaporator for precipitation of the iodine as $Hg(IO_3)_2$.

---

*This discussion covers the process as developed in the United States. Although work on this process has also been done in Europe, the status of that work was not investigated.

This process has been studied on a laboratory scale (30 liter/m or ∿ 1 ft^3/min) at ORNL and at Savannah River, where elemental iodine DFs (at optimum Hg concentrations) of 2260-8400 and methyl iodide DFs of 10^4 and above were obtained.(12) The higher I_2 DFs were at higher concentrations of HNO_3, but higher nitric acid concentrations decreased efficiencies of CH_3I removal. (This confirmed similar results at ORNL.)

At Savannah River, a full-scale iodine control process is also in operation using dilute solutions and DFs below the commercially acceptable level. At the Allied Gulf Nuclear Services (AGNS) plant at Barnwell, S.C., a Mercurex-type process has been installed that is conceptually similar to a system used for iodine removal at a recovery facility in Idaho. The Idaho process was run for several years at DFs comparable to those expected at the AGNS plant (ten per scrubber, two scrubbers in series). DF for elemental iodine is expected to be higher than 10; for CH_3I, less than 10.

The main disadvantage of the Mercurex process appears to be a high mercury use (4:1 molar mercury to iodine), translating to 30 m^3 of liquid waste/yr(1) and lack of commercial scale demonstration of either the process or a solidification/recycle process system. The waste problem is caused by high iodine carryover as the retained iodine builds up in the scrubbing solution. The iodine carryover is in a form (unidentified) not readily removed by a second Mercurex train in series. A solid adsorber or Iodox tower will apparently remove this iodine form. In order to minimize this carryover, mercuric iodide must be removed in large quantity to maintain system efficiency.

The information available regarding testing of the Mercurex process indicates that there is still some question regarding its tolerance to potential impurities in the off-gas (substances other than O_2, N_2, Ar, H_2O). The AGNS off-gas control system provides for NO_x absorption between the two iodine scrubbers; plans were to move this upstream of the iodine removal system if NO_x interferences in iodine removal required it.(2a.)

B-III.2.3 Iodox

The Iodox process uses a highly concentrated nitric acid scrub process in a bubble-cap tower to oxidize iodine to nonvolatile iodate (organic iodine is oxidized to free iodine first). The resultant solution is evaporated to a solid and results in a very low waste volume (0.4 m^3/yr).(11) The Iodox process has been developed at ORNL through a pilot-scale plant. DFs as high as 3 x 10^4 were reported for seven plates at 30 liter/min (1 standard ft^3/min) (average DF of 4.4 per plate). Groenier and Hannaford concluded that, "treatment of a 100 ft^3/min off-gas stream to obtain a DF of 10^5 could theoretically be accomplished using a 20-inch diameter bubble-cap column having 10 plates."(13) (DF of 3.2 per plate.) The pilot-scale tests used a 7.6-cm (3-inch) diameter bubble-cap column. A more conservative estimate has been that a DF of 10^4 is achievable.(1)

Work has also been performed with packed towers, but the bubble-cap process was stated to be preferred due to better control over a range of flow rates. Results for CH_3I varied from no change from I_2 efficiencies to lower than I_2 by as much as a factor of ten. High CO_2 levels did not significantly affect results. Presence of certain organics (hexane, notably) reduced results by a factor of five.

Apparently Iodox results on a pilot-scale were very encouraging. The main disadvantages of the Iodox process are the highly corrosive scrub liquid and the fact that nitrogen oxides (NO_x) interfere with the process by shifting the chemical equilibrium (this is not a severe problem at a few percent NO_x and, in any event, is a system design problem, not a fundamental one); the corrosion problem can probably be dealt with satisfactorily (according to sources at ORNL, the U. S. Army has considerable experience with high-concentration HNO_3 in stainless steel and Zircaloy). The fact that the Iodox process has not operated on a commercial scale makes the success of its application to a reprocessing plant uncertain. The Iodox process (with solid adsorbent polisher) has been selected for conceptual design of an LMFBR reprocessing system.

ORNL has recently been studying use of electrolytic scrubbing for removal of iodine.(14) The principal advantage to this method is that lower-concentration nitric acid may be used. Tests were performed on a laboratory scale (1350 ml/min) with sample gas containing air, NO_x, water vapor, and iodine (both elemental and organic). At about 100 ppm (vol) CH_3I, DFs of about 100 were obtained. For elemental iodine, 20-40 ppm, DFs of about 600 were obtained. Best results for both I_2 and CH_3I appeared to be at $8\underline{M}$ HNO_3; DFs ranged from 111 (2% NO present) to 685 for I_2, 5.6 (2% NO present) to 113 for CH_3I. The results reported suggest that NO in 1-2% quantities severely degrades the DF.(14) NO_2 also degrades performance: 1% NO_2 lowered CH_3I DF from 100 to 10, and I_2 DF from 600 to about 150. Electrochemical reduction yields several reduction products: NO, N_2O, H_2, and ammonia.

Several methods of concentrating and storing iodine products from the electrolytic process are being investigated; although there are no conceptual problems with distilling off the nitric acid to leave iodine residue, the technology has not been proved.

B-III.2.4 Chemisorption

Laboratory tests using silver-loaded zeolite (AgZ) have yielded DFs of 10^2 - 10^5 for I_2 and organic iodine in air streams. Tests at Idaho National Engineering Laboratory (INEL) using NO_2 and H_2O contaminants yielded DFs reduced by 2-3. The chemisorption technique has wide support because of its simplicity and ease of waste handling; however, it is included in conceptual designs mainly as a "polisher" because of concern over the high cost of silver.(6) Annual silver cost for removal of 600 kg (21,120 oz) of iodine at 25% efficiency and $5/oz is about

$$(21{,}120)\ (\$5)\ \frac{(108)}{(128)}\ \frac{(1)}{(0.25)} = \$356{,}000$$

This is a possible alternative to more complicated, high-capital-cost systems. Some concern has also been voiced regarding heat-removal problems in large, commercial-scale beds, although for LWR fuel reprocessing, this would not be a problem because of the low residual amount of radioiodine after a few months of fuel cooling.

The results of tests of AgZ at Savannah River with I_2 and CH_3I in a 7 liter/min air stream showed a DF of 10^4 for I_2, 250 for CH_3I.(12) For PbZ a DF of 333 was achieved for I_2 but only 1.8 for CH_3I. Other forms tried were less effective. Yearly waste volume is expected to be about 5.7 m^3 and this amount is expected to be satisfactory for long-term storage if NO_2 and moisture are purged.(1) The cost of AgZ adsorbent has been estimated at 3-5 times higher than that for the Mercurex process.(1)

Because of the high cost of silver, methods are being evaluated for regeneration of the AgZ bed into a PbZ bed, with a hydrogen stream used to transport the I as HI between beds. Also, system design could reduce silver requirements by improving efficiency (e.g., going beyond 1:4 iodine loading, providing polisher beds in series).

Other than the data from testing of silver zeolite beds with NO_2, and water-saturated air streams (with no serious degradation of efficiency), no data are available regarding performance of this treatment technology with regard to other impurities (CO_2, solvent carryover).

B-III.2.5 Conclusion

There is no tested technology on a commercial scale in the United States for removal of iodine at high DFs from the off-gas stream of an LWR reprocessing plant. Caustic scrub will probably never be used because of low efficiencies. All of the remaining techniques--Mercurex, Iodox and adsorbent--have been tested at laboratory- or pilot-scale levels, and all have been shown to have potential for iodine removal, with Iodox demonstrating the best characteristics for removal of both elemental and organic iodine. Current system designs, Allied General Nuclear Services (AGNS) and LMFBR conceptual design, have either Mercurex or Iodox front-end scrubbers followed by an AgZ polisher. Both techniques appear to be feasible.

B-III.3 CARBON-14

B-III.3.1 Background

The U.S. Environmental Protection Agency has been considering the need for control of carbon-14 (C-14) effluents from the nuclear fuel cycle.(15,16) For orientation, it may be useful to compare the amount of

C-14 to be evolved from the LWR fuel cycle with that present as a result of natural processes.

C-14, a radioisotope of carbon with a half-life of 5700 years, is being produced continuously in the upper atmosphere by nuclear changes in stable N-14 (caused indirectly by cosmic rays). Enough is produced to support a relatively constant inventory of about 4.5 MCi (megacuries) of C-14 in the atmosphere worldwide. This C-14 (about 1 MT) is interspersed uniformly in the carbon contained in the CO_2 of the atmosphere, and ultimately in the carbon contained in every living thing, at a C-14 level of about 15 disintegrations/minute (d/m) per gram of total carbon.

When an organism dies and ceases to exchange with the atmosphere (by photosynthesis, breathing, or eating of other life forms), its C-14 level drops, with a 5700-year half-life. This provides the basis for the so-called radiocarbon dating technique used for determining the age of ancient carbon-bearing relics by comparison of their present C-14 levels with an assumed value of 15 d/m per gram of carbon at the time of their death in the past. Recent calibrations of this technique with wood from the individual tree rings of bristlecone pine trees (thousands of years old) show, however, that the C-14 abundance in the carbon of the atmosphere (and thus of living things) has not always been constant; 4000 years ago, it was 6% higher than it is today, and 7000 years ago, it was 10% higher than it is today. These changes may be associated with changes in the earth's magnetic field.(17)

The total amount of C-14 that would ultimately be emitted, if uncontrolled, from the operation of the 700 GW of U.S. reactor capacity (see Task A Report) to be built by the year 2020 would be, at 28 Ci/GW-yr (see below) about 700 x 28 x 30 = 0.6 MCi of C-14. If 40% of the C-14 emitted stays in the atmosphere,(15) and if the world total is triple the U.S. figure, then the atmospheric inventory would, when all those reactors shut down by 2040 or so, have risen by 3 x 0.6 x 0.4 or 0.7 MCi of C-14, or 16% of the natural C-14 background. Since the combustion of fossil fuels (which have no C-14 left in them because of their great age) should simultaneously increase the CO_2 content of the atmosphere by several percent of the present level, the level of C-14 per gram of carbon would rise by less than the maximum figure given above.

If one assumes a rise of 16% in the nominal human body content of about 85 nCi of C-14, this should result in an additional annual personal dose of about 0.2 mrem, or about 1/500th of the present total dose from all sources, or an increment equivalent to that received from cosmic radiation in one hour of commercial air travel.(18)

Thus, as shown above, even 60 years from now, the effect on personal background radiation would still be: a) at most a very small quantity, and b) of the same order as that caused by previously-seen naturally-occurring changes in C-14 levels. Although there are unresolved questions regarding the need for C-14 control in the near future, the following discussion presents the current state of knowledge concerning C-14 control technology.

Quantities of C-14 are produced both at fuel reprocessing plants and also at nuclear reactors. The major source of C-14 in nuclear fuel probably comes from the (n,p) reaction with N-14, which is present as an impurity in fuel; concentrations are of the order of 10 ppm, and some values as high as 25 ppm have been reported.(1) Production of C-14 in reactor coolants is primarily from the (n,α) reaction with O-17 (which occurs in natural oxygen with an abundance of 0.037%).(16) Although other possible reactions mentioned are C-13(n,γ), N-15(n,d), and O-16(n,He-3), the N-14(n,p) reaction in fuel and the O-17(n,α) reaction in coolant are believed to be the most significant sources of C-14.(1) Breakdowns of C-14 source terms have been estimated as follows:(16)

Fuel

N-14(n,p)C-14 → 18 Ci/GW-yr (assuming 20 ppm N-14 by weight in fuel)
O-17(n,α)C-14 → 4 Ci/GW-yr (from 0.037% O-17 in the UO_2)

BWR

O-17(n,α)C-14 → 8.9 Ci/GW-yr (from oxygen in coolant)
N-14(n,p)C-14 → 0.26 Ci/GW-yr (assuming 1 ppm N_2 impurity in coolant)

PWR

O-17(n,α)C-14 → 3.3 Ci/GW-yr
N-14(n,p)C-14 → 0.09 Ci/GW-yr

Thus, according to these estimates, approximately 22 Ci/GW-yr are produced from fuel, and would appear as reprocessing plant effluents, and approximately 3.4 and 9.2 Ci/GW-yr would be released from PWR and BWR reactor plants, respectively, (average of 6.3 Ci/GW-yr if it is assumed that reactors are equally distributed between PWRs and BWRs). These numbers indicate that although reprocessing plant releases will account for the majority of C-14 discharged from the fuel cycle, releases from reactor plants are also significant and, if C-14 is deemed to be worth controlling, these releases will also have to be dealt with. Table B-III-1 presents a range of estimates of C-14 production rates.(16)

Actual measurements taken at a BWR(19) showed gaseous releases of 16 Ci/3,000 MW_t-yr, which is equivalent to about 16 Ci/GW-yr. These studies showed the C-14 to appear mainly as CO_2 (95%), with the remainder being CO and hydrocarbons. Similar studies at a PWR(20) showed 8 Ci/GW-yr, of which 80% were hydrocarbons (CH_4, C_2H_6) and less than 5% were CO_2 and CO.

It has been estimated(1,8) that the maximum concentration of $^{14}CO_2$ in the dissolver off-gas is approximately 0.025%; CO_2 in the off-gas is approximately 0.03% (CO_2 concentration in air). Thus, the maximum $^{14}CO_2$ concentration would almost equal the natural CO_2 concentration. This estimate varies substantially from other estimates(16) that about 0.02%

TABLE B-III-1

PRODUCTION OF CARBON-14 IN LIGHT-WATER REACTORS

		Carbon-14 Production Rate (Ci/GW-yr)				
		ORP/TAD 76-3 (16)	Bonka et al. (21)	Hayes et al. (22)*	ERDA-1535 (23)	Kelly et al. (24)
BWR Fuel	0-17	4	8.4	10.9		2.7
	N-14	18	12.9	21.2		10.9
	Total	22	21.3	32.1	20**	13.6
BWR Coolant	0-17	8.9	9.9	11.5		
	N-14	.26	1.3	--		
	Total	9.2	11.2	11.5	16	16
BWR Sum (Fuel Plus Coolant)		31	32.5	43.6	36	29.6
PWR Fuel	0-17	4	7.1	4.0		2.7
	N-14	18	12.2	7.6		10.9
	Total	22	19.3	11.6	17**	13.6
PWR Coolant	0-17	3.2	9.8	3.3		
	N-14	.09	1.3	0.1		
	Total	3.3	11.1	3.4	6	6
PWR Sum (Fuel Plus Coolant)		25	30.4	15	23	19.6

*The production rates presented by Hayes et al.(22) and Kelly et al.(24) for 1000 MWt were multiplied by 3.03 (33% thermal efficiency to roughly present the values on a per GWe-yr basis for comparison purposes.

**Fuel and cladding production rates from ERDA-1535(23) were added and identified as a fuel production rate in this table.

Source: Public Health Considerations of Carbon-14 Discharges from the Light-Water-Cooled Nuclear Power Reactor Industry. ORP/TAD-76-3, U.S. Environmental Protection Agency, July 1976.

of the CO_2 flow is expected to be $^{14}CO_2$. The latter estimates(16) have been checked and appear to be reasonable: an air flow rate of 1.18×10^6 g/hr is given, which equates to 15.1 m^3/min (550 ft^3/min) estimated in ERDA 76-43.(1)

$$\frac{(1.18 \times 10^6\ \text{g/hr})\ (0.035\ \text{ft}^3/\text{liter})}{(1.29\ \text{g/liter})\ (60\ \text{min/hr})} = 534\ \text{ft}^3/\text{min or } 15.1\ \text{m}^3/\text{min}$$

ORP/TAD-76-3 gives $^{14}CO_2$ flow rate of 0.017% of the CO_2 flow rate of 592 g/hr.(16) This equates to 20 Ci/GW-yr:

$$\frac{(0.00017)\ (592\ \text{g/hr})\ (7200\ \text{hr/yr})\ (1.37\ \text{Ci/g}\ ^{14}CO_2)}{50\ \text{GW}} = 20\ \text{Ci/GW-yr}$$

This is compatible with other estimates within the range of uncertainty, supporting the estimate in ORP/TAD-76-3.

The concentration of $^{14}CO_2$ in CO_2 is not important from the standpoint of suitability of removal technology; what is important is the concentration of CO_2, and the removal technology selected must be capable of reducing the 0.033% of CO_2 in air (0.07% by some estimates if dissolver additions are considered and a very low air sparge is used) down to a level which will allow effective removal of C-14.

Estimates have been made that from 95-99% of the C-14 in the nuclear fuel will be released at the reprocessing plant dissolver as CO_2 or CO with the remainder "likely to be incorporated into carbonic acid".(16) There do not, however, appear to be any empirical data supporting this assumption, although it is a reasonable one in view of the conditions at the dissolver: 90°C, 8 M nitric acid, and excess O_2 in solution from the UO_2 and HNO_3.(16) The 95-99% figure should not be relied on as definite. The fate of the 1-5% of C-14 remaining is uncertain but ORP/TAD-76-3 states that most likely it will be discharged with the high-level waste stream.(16) A third source of uncertainty is how much C-14 will form CO_2 and how much will form CO. This is an important point, because significant ^{14}CO will require the installation of a $CO \rightarrow CO_2$ converter--probably not a difficult problem, but one that must be accounted for.

The following discussion of methods of CO_2 removal from dissolver off-gas streams is narrowly focused on the assumption that significant C-14 will appear as CO_2 in the off-gas to make control of this CO_2 the main problem; this assumption is a reasonable one but ORP/TAD-76-3 warns:

> "In summary, the need for research on C-14 in reprocessing facilities exists in the following areas: amounts of C-14 produced in the fuel, chemical forms of the C-14 as it is evolved from the dissolver, unexpected chemical behavior elsewhere in any system, partitioning between the off-gas and liquid process systems, possible process pathways and

reactions in those systems , probable decontamination factors for removal systems, and costs of collection systems."(16)

Three systems have been identified for possible use in removal of CO_2 from the dissolver off-gas system: caustic scrubbing, molecular sieve adsorption, and fluorocarbon absorption.(7) The first two have been used for CO_2 control in other applications but not specifically for dissolver off-gas application; the third method has been demonstrated on a small scale for Kr-85 control from simulated dissolver off-gas (see Section on Kr-85 control).

B-III.3.2 Caustic Scrubbing

This technique for removal of CO_2 from an air stream depends on the reaction of hydroxides such as NaOH with acidic gases (CO_2, NO, NO_2) to produce carbonates and nitrites. The system block diagrams shown in ERDA 76-43(1) provide a two-stage scrub: first a pump utilizing NaOH as a working fluid and sealant, then a caustic spray tower. By addition of calcium hydroxide ($Ca(OH)_2$) to the stream, $CaCO_3$ (calcium carbonate) is precipitated out. This calcium carbonate may then be dried and packaged.

ERDA 76-43 suggests that annual output of $CaCO_3$ would be 16-60 kg,(1) but this appears to be in error. As shown previously, approximately 592 g/hr of CO_2 is generated, which equates to:

$$(592 \text{ g/hr})\ (7200 \text{ hr/yr}) = 4262 \text{ kg/yr of } CO_2\text{, which translates to:}$$

$$(4262)\ \left(\frac{100 \text{ g } CaCO_3}{44\text{g } CO_2}\right) = 9686 \text{ kg/yr } CaCO_3$$

a much larger quantity than stated in ERDA 76-43, but still within a range that may be easily managed for ultimate disposal.

Use of caustic scrubbing to remove CO_2 from air is a common industrial application. However, the process has never been tested for dissolver off-gas application, and, therefore, the suggestion(1) that 99% removal rate may be achieved is speculative.

B-III.3.3 Molecular Sieve Adsorption

In this process, the above-described caustic scrub steps for isolation of the CO_2 stream are replaced by adsorption on Type 5A molecular sieve. This is accomplished by removal of NO and NO_2 with dilute HNO_3, destruction of N_2O on a rhodium catalyst, drying on molecular sieve 3A, and CO_2 removal on molecular sieve 5A. In order to collect the carbon, it is proposed that the molecular sieve be regenerated (by heating in a vacuum or a gas purge), then the regenerate gas scrubbed to produce $CaCO_3$.

As with caustic scrubbing, the above-described technology (except acid-circulating and sealed pumps) has been demonstrated or is conceptually straightforward; the difficulty lies in that no integrated system for dissolver off-gas application has been demonstrated.

B-III.3.4 Fluorocarbon Absorption

The section on krypton control discusses fluorocarbon absorption at length and points out that other gases besides krypton will be absorbed by R-12, including CO_2, and that pilot facility tests showed CO_2 decontamination factors of over 10^4. The stripper off-gas from the krypton-oriented process contains Kr, Xe, Ar, and CO_2. How the CO_2 would then be removed from the product gas is not clear, since storage as a compressed gas is not suitable because of the long half-life of C-14. Caustic scrubbing could be used to yield calcium carbonate, presumably at some saving over direct scrubbing of the off-gas, since smaller volumes of gas are involved after the absorption process.

The fluorocarbon system has been tested conceptually and found to be effective at CO_2 removal, although final CO_2 removal/isolation has not been tested. (The fluorocarbon system is explored in more detail in the discussion of krypton.)

Figure B-III-2 summarizes the three systems.(8)

B-III.3.5 Conclusions

The requirement for control of C-14 from the nuclear fuel cycle by EPA appears possible. A large percentage (about 20%) of C-14 production from the fuel cycle comes from power reactors, and, therefore, meaningful control of C-14 could be accomplished only by control at reactors as well as at reprocessing plants. Because reprocessing is not being done at present in the United States, reactor plant off-gas technology will likely provide a much greater technical base for C-14 control than presently exists and may provide this base before reprocessing plant control is required, thereby considerably easing the technical uncertainty for U.S. reprocessors.

Some uncertainties remain in the areas of C-14 production sources, rates, and chemical form from the reactor and from the reprocessing plant. These questions must be answered before an effective control technology can be developed. Once the sources, rates, and forms of carbon that must be controlled are more clearly understood, flowsheets may then be developed on the basis of these quantified factors. Questions must then be answered regarding acceptability of each proposed method for the postulated effluent stream composition and percentage content of CO_2. If substantial carbon forms other than CO_2 are present, either alternate processes must be provided, or these forms must be converted to CO_2.

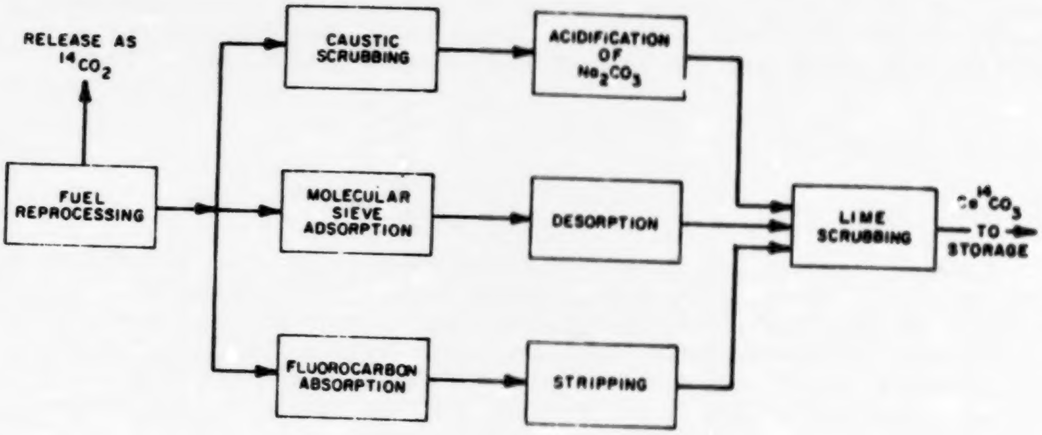

RELEASE AS $^{14}CO_2$
FUEL REPROCESSING
CAUSTIC SCRUBBING
ACIDIFICATION OF Na_2CO_3
MOLECULAR SIEVE ADSORPTION
DESORPTION
FLUOROCARBON ABSORPTION
STRIPPING
LIME SCRUBBING
$Ca^{14}CO_3$ TO STORAGE

FIGURE B–III–2 ALTERNATIVE TECHNOLOGIES FOR THE MANAGEMENT OF C–14

Source: Proceedings of the International Symposium on the Management of Wastes from the LWR Fuel Cycle. CONF-76-0701, U.S. Energy Research and Development Administration, July 1976.

Three technologies have been identified by ERDA as possible candidates for commercial application at fuel reprocessing plants for removal of CO_2 from dissolver off-gas streams: caustic scrubbing, molecular sieve adsorption, and fluorocarbon absorption. None of these systems has been proved for application to reprocessing plants, although CO_2 removal from air by caustic scrubbing is a common commercial process, and insofar as reprocessing plant application is similar to removal from air, the technical problems will be minimized.

Finally, the possibility should not be overlooked that the most cost-effective method of C-14 control could be reducing the nitrogen content of the fuel during the fabrication process. No definitive work appears to have been done in this area, nor is it expected that any would be done until the C-14 control question has been better defined.

B-III.4 TRITIUM

B-III.4.1 Background

Current reprocessing plans call for release to the environment of essentially all of the tritium produced at a fuel reprocessing plant. There are no absolute limits on tritium releases from the fuel cycle, but tritium releases must meet effluent concentration limitations set by the Nuclear Regulatory Commission in 10 CFR 20. Controls to meet concentration limitations do not require absolute retention, but rather dilution.

It is estimated(1) that more than 98% of the tritium produced in LWRs (from ternary fission, with a fission yield of approximately 10^{-4}).(6) will remain in the fuel to the reprocessing plant. Because of the low production rate, however, the tritium will be present in very dilute quantities; concentration in the dissolver is estimated to be about 2 ppm.(6) Estimates of curie-quantities of tritium vary according to the fuel design (plutonium fission produces more tritium than uranium fission).(25) Values vary from 415 Ci/MT of fuel(25,2a) to 490 Ci/MT(26) to 700 Ci/MT.(27) Under existing fuel reprocessing plant designs, control of this tritium is impossible except by collection and storage of large volumes of waste water.

The distribution of tritium in a fuel reprocessing plant is not well understood, as witnessed by large variations in the estimates in various references. The following estimates have been given for tritium pathways.

Substantial tritium will remain in the hulls, estimates ranging from 10%(25) to 10-50%.(1) Some tritium will be released in the dissolver off-gas as HT, estimates ranging from very little (1%(1)) to substantial (10%(25)). During a zirconium fuel processing campaign at ICPP, 10-45% of the contained tritium was released from the dissolver as HT.(27) Another estimate is that 20% will leave at the dissolver.(28) The remaining tritium will remain in the aqueous streams and, unless controlled, will ultimately be discharged to the environment in liquid waste streams or as vapor. The uncertainty regarding distribution was discussed by Musgrave, who concluded:(27)

> "Data are extremely sparse on the chemical state and distribution ratios for tritium in any of these streams. While the majority of the tritium will eventually appear in effluents as the oxide or element, its combination with organic materials will present many additional control problems if release to the environment is to be completely avoided."

This organic exchange (which transfers the tritium from the first extraction cycle contactor to the low-level areas of the reprocessing stream) was estimated to be minimal (less than 1%).(25,26) However, Hall and Ward considered this to be a significant pathway in recycle schemes:

> "...The uranium-plutonium bearing organic stream...also transfers 0.004 g/l water and 0.14 molar nitric acid across the imaginary isolation boundary separating the two process sides. This stream, an unavoidable cross-over path from the leacher-feed solution to the low level process side, accounts for 6 to 20% of the daily tritium input being transferred across the boundary [in the reference recycle scheme]."(29)

Given the above unknown regarding the precise behavior of tritium at fuel reprocessing plants, it is not surprising that considerable differences exist in proposed means of dealing with tritium. These tritium-control proposals fall into two broad categories: head-end processes (voloxidation and pyrochemical techniques) and process-stream controls (recycle and/or isotopic separation). A third possibility, retention of the entire water effluent (including, presumably, the water removed from gas streams) seems practical (economic) only if an on-site geologic method, such as dumping into deep, stagnant formations, can be used.

B-III.4.2 Voloxidation

The voloxidation process would provide a front-end kiln to heat chopped fuel elements to drive off tritium and some other fission products and oxidize UO_2 to U_3O_8. Testing of the voloxidation process(30) has shown that over 99% of the tritium will become volatilized (as HTO) at temperatures ranging from 450-650°C. Conceptual design using the voloxidation process(1) envisions passing off-gas from the chopper (tritium would be emitted as HT) through an oxidizer (Ni-Cr-Pd catalyst, 400-600°C) to convert HT to HTO, then joining with the voloxidizer off-gas and cooling to condense the HTO, which may then be removed by a drier-molecular sieve arrangement. The off-gas stream, which would contain large quantities of noble gases and iodine, would then be routed to the off-gas system for treatment. Murbach et al. report a different flowpath, joining the chopper/voloxidizer off-gas upstream of the catalytic oxidizer.(6)

Substantial problems and uncertainties exist with the voloxidation concept. Testing has shown that $UO_2 \rightarrow U_3O_8$ conversion is not always complete, depending upon residence time, kiln rotational speed, oxygen content of the feed gas, and fuel pellet composition and fabrication technique.(31) Testing has shown consistently good and predictable results on tritium removal, however, indicating that 99% release may be readily achieved in the 450°C area. Below 350°C no oxidation occurs.(30,31)

One of the most troublesome problems with voloxidation is the behavior of other fission products. Tests have shown that the process releases krypton and xenon, and volatilizes ruthenium, cesium, cerium, antimony, and niobium, as well as significant quantities of uranium and plutonium. The release of extraneous fission products is not complete. Goode suggests that about 50% of the krypton/xenon will be released from the shearing step, and an additional 25% will be released by voloxidation.(30) Iodine removed by voloxidation varied from 40% at 450°C to 74% at 650°C (higher than 650°C temperatures reduced iodine release). Thus, at the likely operating temperature of 450°C, less than one-half the iodine will be emitted at the voloxidizer. Release of other elements was investigated by heating fuel rod segments to 450°C for 3 hr, then 750°C for 7.4 hr. The following releases occurred: Nb-95, 9.4%; Ru-106, 43.2%; Sb-125, 25.8%; Cs-137, 29.1%; Ce-144, 3.3%; U, 2.2%; Pu, 3.4%.

The release of xenon and krypton is probably not a problem, since these inert gases will simply pass through the tritium recovery system and into the off-gas control system. The other elements, however, are a very significant potential problem, because of their tendency to plate out at cold spots. Testing showed that most of the elements released (other than inert gases) plated out either on the burner walls or burner head, or were trapped in the molecular sieve. Because of the volatility and subsequent plateout of fission products, there are significant questions concerning maintainability and waste handling aspects of the voloxidation process. Clearly, the system must be absolutely leak-tight, at least to the molecular sieve.

Another question that arises from the volatility-plateout question covers iodine control. Meeting the 99.6% retention requirements of 40 CFR 190 for I-129 will certainly be complicated if some of the iodine is left in the voloxidation system.

A final question regards safeguards requirements. Present U.S. regulations (10 CFR 70) require periodic physical inventories to verify the plant material balances to within 1% for plutonium and 0.7% for U-235. Such requirements may not be met if 2-3% of the uranium and plutonium are retained in the voloxidizer. Physical location and assay of this uranium/plutonium would be virtually impossible without major disassembly of the highly contaminated voloxidizer system.

Other potential voloxidizer problems have been listed by ERDA 76-43:(1)

1. The $UO_2 \rightarrow U_3O_8$ oxidation is exothermic and a difficult control problem results.

2. Handling the U_3O_8, a fine powder, will present difficulties.

3. Nuclear criticality considerations (although this is listed as a potential problem, it does not appear to be any more difficult than criticality considerations in other transfer systems).

4. Cleaning and maintenance of the voloxidizer system.

5. Potential for thermal failure of the high-temperature kiln.

In addition, there is the hazard of possible ignition of the Zircaloy cladding and fines by the high-temperature oxidizing gases.

Although extensive data from laboratory-scale and hot-cell tests demonstrate the fundamentals of the voloxidation process (and identify some of the potential problems), engineering development has been limited to tests with unirradiated UO_2 in a rotary calciner.(30) A tritium-control flowsheet has been developed for the AGNS plant(6) along with a cost estimate of $34 million in 1973 dollars.

At Savannah River a series of laboratory-scale tests are planned (starting January 1977) of head-end tritium removal technologies,(26) which will involve voloxidation of irradiated LWR fuel rod sections. These tests will include trapping of off-gases in order to determine the composition of the streams. These experiments should provide substantial additional data for evaluating the voloxidation process.

Estimates of waste quantities from the voloxidation method are less than 100 liters/day for a 5 MT/day plant.(8)

B-III.4.3 Pyrochemical Processing(1)

Pyrochemical processing consists of first decladding the fuel by selective melting of the cladding. With stainless steel, this is feasible (melting point, 1450°C), but with zirconium-based cladding, the required melting point (1840°C for zirconium) may be too hot. UO_2 melts at about 2100°C. Following the cladding melt, the fuel is reduced in a solution of reductant alloy (Zn, Ca, Mg) and a salt (e.g., $CaCl_2$) at 800-900°C. Salt is recovered and recycled, and the reductant alloy is distilled off for recycle, leaving U and Pu to be sent to the acid dissolver. An alternative process is to precipitate the U and Pu through addition of calcium nitride.(1)

Fission product tritium and noble gases are both released during decladding and reduction phases. Iodine is thought to remain in the salt phase,(1) but this seems highly speculative in view of the high temperatures required. Fission product volatility is stated to be a potential problem.

Most work done on pyrochemical processing has been done at Argonne National Laboratory, and has been directed toward the LMFBR. According to ERDA 76-43, "although considerable development work has been carried out involving various concepts, a complete process has not been demonstrated for LWR of individual processes that appear to have potential in a Purex head-end step."(1)

Because of the considerably higher temperatures required in the pyrochemical process than in the voloxidation and the substantially more complicated chemical systems, voloxidation will probably prove to be more favorable than pyrochemical methods.

B-III.4.4 Process-Stream Treatment

Two techniques have been proposed for tritium control once it enters the aqueous streams at a fuel reprocessing plant: isotopic separation and water recycle. These two concepts should correctly be discussed together, since isotopic separation is normally considered to be supplementary to recycle.

At one end of the recycle-separation spectrum is total recycle without separation. In this case, tritium will build up to a high-concentration equilibrium (addition from fuel equal to removal by waste streams and decay). At the opposite end of the spectrum is no recycling but separation of tritium from the effluent stream. A middle ground, and the most favored option, is recycle with bleedstream separation to keep in-plant concentrations tolerable.

The obvious problem with total recycle without separation is that tritium concentrations eventually become sufficiently high to present severe in-plant safety problems. Even a small leak could be quite serious to operating personnel. This results in the need for costly gas control systems. On the other hand, the problem with no-recycle separation of tritium from the effluent stream is that existing processes for tritium separation from water are uneconomic at high-volume flowrates. Likewise, storage of the entire aqueous waste stream would be uneconomic without some degree of tritium concentration to reduce storage volume.

General Electric (GE) has performed an important study of tritium control by water recycle,(29) in which seven cases of recycle process options were examined and material balances were established for each case in order to determine the probable tritium distribution and effectiveness of tritium retention. The process systems were, for simplicity, divided into three segments: the leacher (dissolver), the high-level system, and the low-level system (determined by total radioactivity, not just tritium). This tritium equilibrium model is shown in Figure B-III-3. The seven cases are briefly described in Table B-III-2, and the resultant balances are given in Table B-III-3.

Recycle design considerations demand extensive recycle of water and acid, removal of water from vent gases, extensive personnel protection efforts (seals, containment, ventilation control), and avoidance of contaminated gas contact with hydrophilic substances such as concrete.

GE's reference recycle scheme, Case 1, utilized a modified Purex process and is arranged to maximize the use of recycle water throughout the plant and minimize aqueous inputs. The recycle water systems are segregated into a high-activity system (feed preparation, HA wastes) and a low-activity system serving the remainder of the process building. As may be seen in Figure B-III-3, tritium output is from two points only: leacher off-gas, and tritiated wastes from the LA system. Table B-III-3 shows that Case 1 results in high LA concentration relative to HA. This is undesirable and is changed in Case 2 by removal of all of the tritium from the LA side, which reduces concentrations on both sides of the process. In Case 3, new acid is added to the LA side only, the addition resulting in further dilution of the LA side but pushing the HA side back up. Case 5 combines Cases 2 and 3, but no significant improvements are realized.

Case 7 turns out to be the most advantageous, for here all aqueous crossover from HA side to LA side is eliminated, and the only transfer is through the organic stream from the high-activity column. The tritium concentration on the low side is 0.63 Ci/liter, the lowest of all seven cases. However, even with the relatively low tritium concentration, 0.63 Ci/liter, the report estimates that: "this degree of tritium isolation would probably not be sufficient to significantly reduce the tritium concentration in the low level process side to the extent that the personnel protection...could be substantially reduced."(29)

Although cost determination was not a goal of the GE study, an estimate was made that recycle would increase plant cost by 20-25%.

The GE study was selected as a base from which to discuss recycle because it points out several very important points regarding recycle:

1. Contrary to some studies, it may not be reasonable to expect near-complete tritium isolation to the high-level side. The GE study suggested that personnel protection problems would

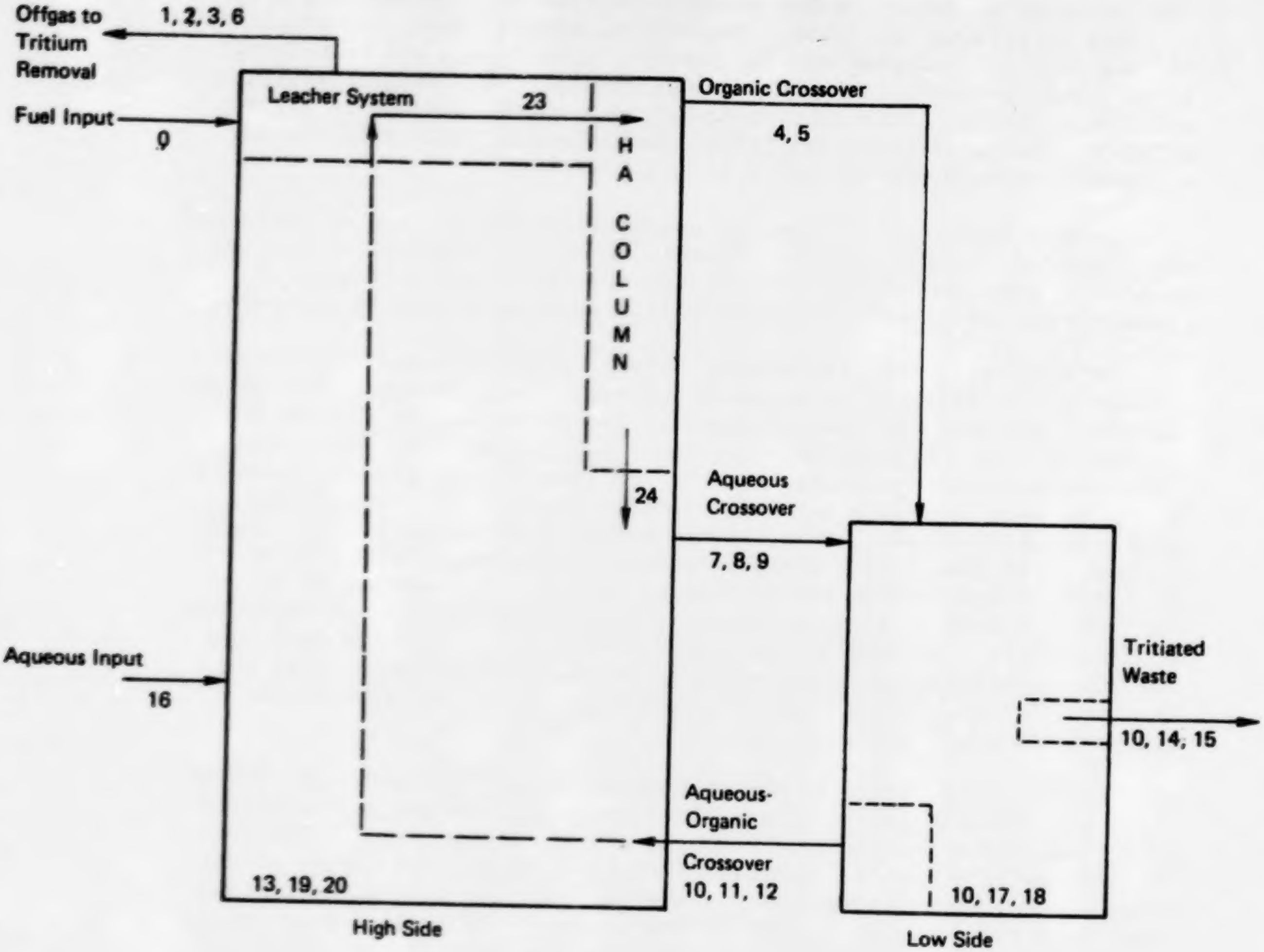

Offgas to
Tritium
Removal
1, 2, 3, 6
Fuel Input
9
Leacher System
23
H
A
C
O
L
U
M
N
Organic Crossover
4, 5
24
Aqueous
Crossover
7, 8, 9
Aqueous Input
16
Tritiated
Waste
10, 14, 15
Aqueous-
Organic
Crossover
10, 11, 12
13, 19, 20
10, 17, 18
High Side
Low Side

FIGURE B–III–3 TRITIUM EQUILIBRIUM MODEL

Source: Hall, N.E. and G.N. Ward. Tritium Control by Water Recycle in a Nuclear Fuel Reprocessing Plant. NEDG-11342, General Electric Company, San Jose, California, June 1975.

TABLE B-III-2

DESCRIPTION OF POSSIBLE FLOWSHEET VARIATIONS

Case Number	Deviation from Reference Flowsheet
1	None
2	No tritiated water removed from leacher; all water removed from low level side. Same quantity of water is removed.
3	New acid added to low level side. Equivalent amount of acid is transferred from the low level side to the high level side.
4	Recycle uranium transferred from low level process side to HA feed tank at feed concentration, 1.5 molar U instead of the 2.5 molar U in reference flowsheet.
5	Combination of Case 2 and Case 3.
6	No tritium water removed from leacher; all water removed from low level side. Minimize total water removed.
7	Assumes that tritium is only transferred from the high level side to the low level side by the HAP stream.

Source: Hall, N.E. and G.N. Ward. Tritium Control by Water Recycle in a Nuclear Fuel Reprocessing Plant. NEDG-11342, General Electric Company, San Jose, California, June 1975.

TABLE B-III-3

ANALYSIS OF FLOWSHEET VARIATIONS

Variable Number	Units	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6	Case 7
0	Ci/day	6000	6000	6000	6000	6000	6000	6000
1	Fraction Evaporated	0.065	0.000	0.065	0.065	0.000	0.000	0.065
4	Fraction Carried in HAP	0.0137	0.0137	0.0137	0.0137	0.0137	0.0137	0.0137
8	Liters/day	11884	11884	20344	12726	20343	11884	0
11	Liters/day	1352	1352	23384	2194	23384	1352	1352
14	Liters/day	432	2382	432	432	2382	432	432
17	Total liters	8.8×10^5	8.8×10^5	8.8×10^5	8.8×10^5	8.8×10^5	8.8×10^5	8.8×10^5
19	Total liters	3.5×10^5	3.5×10^5	3.5×10^5	3.5×10^5	3.5×10^5	3.5×10^5	3.5×10^5
6	Ci/liter	1.33	0.95	2.60	1.55	3.28	2.19	2.90
13	Ci/liter	1.13	0.75	2.41	1.35	3.08	1.99	2.70
10	Ci/liter	7.89	2.52	2.13	6.87	2.52	13.89	0.63
3	Liters/day	1950	0	1950	1950	0	0	1950
2	Ci/day	2592	0	5082	3032	0	0	5667
15	Ci/day	3408	6000	918	2968	6000	6000	273
Minimum time to tritium equilibrium (days)		265	206	721	396	874	549	776
Percent daily tritium input released		6.2	2	1.7	5.4	2.0	10.9	0.54

Source: Hall, N.E. and G.N. Ward. Tritium Control by Water Recycle in a Nuclear Fuel Reprocessing Plant. NEDG-11342, General Electric Company, San Jose, California, June 1975.

occur on the LA side even with no water transfer between the two sides, simply due to carryover from the high-activity column.

2. A rather obvious conclusion, but one which is important in understanding the balance between recycle and separation, is that as tritium levels reach equilibrium in the aqueous streams, the concentration in waste streams also increases, allowing for either more economical storage or more economical separation.

3. The conclusion reached by Hall and Ward was that, "based on the findings of this study, tritium control by water recycle in a nuclear fuel reprocessing plant is attainable and appears to merit further study."(29)

A second study(27) ignores the difficulties identified above regarding contamination of the low side but takes the recycle concept a step further and examines alternatives employing concurrent separation.

Isotopic separation of tritium from water streams may be performed with a number of different technologies (discussed later), all of which have the common characteristic of being more economic as volume throughput decreases and as required decontamination factors decrease. Therefore, in a recycle system the higher the allowable tritium concentration in the process streams, the more economic will be the removal.

The first system examined by Musgrave envisions total recycle with periodic removal of the entire inventory to storage, or a bleedstream removal to storage.(27) This concept has already been discussed and probably is not optimal because of the significant problem of protecting in-plant personnel. However, the concept should not be dismissed until the economics of alternative systems have been compared.

The most potentially attractive scheme presented by Musgrave (who also presents a once-through full-stream separation concept, which is probably not economic because of the cost of separation) is operation using recycle. The level of tritium contamination in the process streams would be controlled by removing a bleedstream (low flowrate, high concentration) for separation. Depleted recycle water would then be returned to the plant streams (see Figure B-III-4).

The great potential advantage to this concept is the ability to balance the recycle concentration and separation efficiency to achieve optimum economics. As the tritium concentration in the recycle water increases, the costs of plant maintenance and personnel protection would be expected to increase (probably as a series of step functions). Conversely, as tritium concentration increases, the cost of separation would decrease. It is likely that the optimum cost would lie between the two extremes of no recycle (zero tritium concentration in recycle) to no separation (high tritium concentration in recycle).

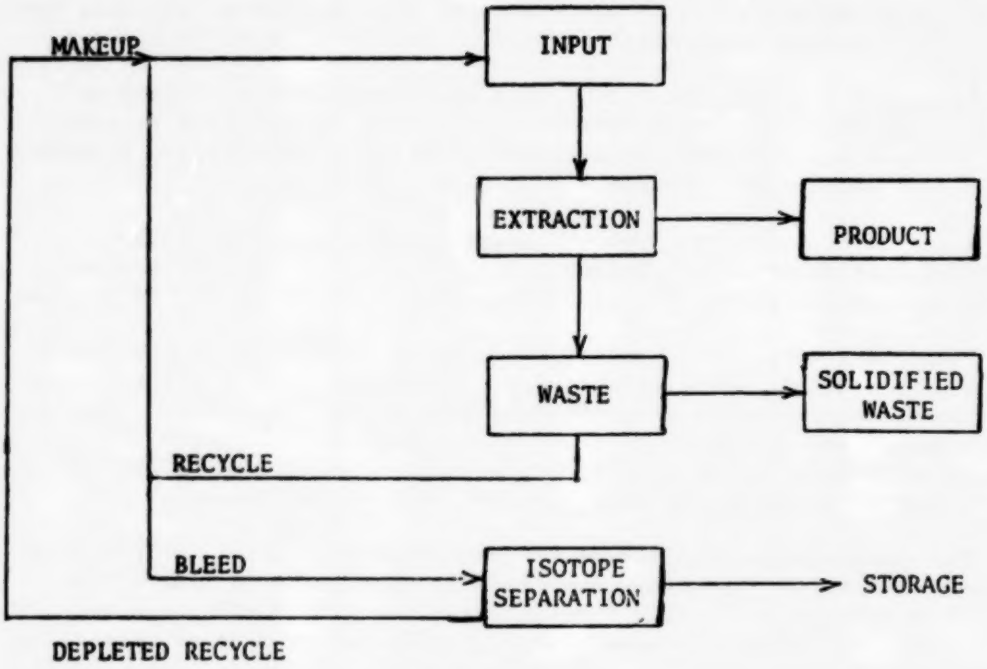

MAKEUP
INPUT
EXTRACTION
PRODUCT
WASTE
SOLIDIFIED WASTE
RECYCLE
BLEED
ISOTOPE SEPARATION
STORAGE
DEPLETED RECYCLE

FIGURE B–III–4 ONE PROPOSED TRITIUM SEPARATION SCHEME

Source: Musgrave, B.C. Tritium Distribution in the Nuclear Industry. ICP-1041, Allied Chemical Corporation, Idaho Falls, Idaho, January 1974.

The above reasoning is consistent with the GE study because as tritium increases in the high level side, it will spill over into the low level side, and this will increase the maintenance/personnel costs accordingly.

The AGNS plant at Barnwell will release all tritium as vapor out the stack.(28) Of the total tritium production rate (estimated at 1.8 x 10^{-2} Ci/sec, or 1555 Ci/day), about 20% will come off at the dissolver, and the remaining 80% will flow with the acid-water mixture from the dissolver, to the high-activity waste concentrator, then to the low-activity waste concentrator, and acid fractionator, issuing in the overheads from the acid fractionator.(28) These overheads will be collected, condensed, sampled, and either recycled or discharged after dilution to acceptable activity levels, if required. Thus, the AGNS design provides for acid-water recycle from the fractionator back to acid makeup, provided tritium contamination is not excessive. In addition, water recycle occurs at several other points in the plant design: GPD receiver, service concentrator feed tank, and 1UD/2UD surge tank.(28, Table 7.5-1)

As already discussed, recycle may cause an increase in tritium concentration in process flowpaths, and this could also be the case at AGNS. This concentration would build up in the water recycle with the acid, and also in tritium exchange with the acid. If the concentration becomes sufficiently high, one might also expect carryover of tritium in the solvent, from water-acid carryover, as well as from tritium exchange with the organic solvent. At AGNS, however, tritium buildup will be avoided by release out of the stack when concentrations become excessive. The expected concentration of tritium in the water discharged from the acid fractionator is lower than that in the GE study(29) by almost five orders of magnitude (8 Ci/liter vs 10^{-4} Ci/liter), a difference suggesting that although water-acid recycle is planned, very little (if any) tritium recycle is expected. How much actual acid-water recycle is feasible with these conditions remains to be seen.

B-III.4.5 Isotopic Separation

Considerable disagreement exists in the published literature regarding the practicality and most promising technology for removal of tritium from water streams. The tritium separation technologies are generally based on heavy-water production techniques and include the following:

1. Catalytic exchange
2. Fractional distillation of water
3. Distillation of hydrogen
4. Electrolysis of water
5. Reversible electrolysis
6. Laser enrichment

Catalytic exchange has been used to produce heavy water on a large scale and thus the method is well understood. The process utilizes the exchange (in the case of tritium):

$$HT + H_2O \rightleftarrows HTO + H_2$$

and occurs when a vaporized mixture is passed over a catalyst. Equilibrium shifts to the right with lower temperatures.

Because the conventional catalytic exchange method requires vaporization of the water, efforts are being pursued to develop an anti-wetting hydrophobic catalyst so exchange may occur between liquid water and gaseous hydrogen in direct contact. The Canadian effort(32) in this field has progressed from the laboratory stage to the engineering stage.

Electrolysis of water is simple and compact(25) and gives high separation factors, but is quite expensive because of high electricity requirements. A technique of reversible electrolysis is being developed(1) that could reduce the power requirements by as much as 80%.(25) Originally, reversible electrolysis was never developed to commercialization possibly because it required expensive palladium membranes. Recently, work in Yugoslavia has produced a technique using a carbon and polyethylene electrode that may be successful for commercial application.

Laser enrichment (by which, theoretically, very high separation factors are possible) is mentioned in ERDA 76-43 as being under development but still at the basic research level.(1) Ribnikar and Pupezin discuss the first four possible separation techniques and favor catalytic exchange or distillation processes from an economic viewpoint.(25)

Of the separation techniques listed, all but laser enrichment and reversible electrolysis have been applied in the production of heavy water and are well understood for this application. However, there is considerable debate regarding their applicability to tritium removal. This disagreement centers mainly on economic questions and ultimately comes to the question of whether an economic mix of recycle and separation may be achieved. There is little question that full-stream (no recycle) separation is uneconomic:

> "The light isotope separation methods, although theoretically possible, are not practical. For example, over 1 tonne per hour of water is vaporized in the waste concentrator. The requirements to electrolyze this quantity of water to obtain the mixture of hydrogen isotopes for separation is impressive. Further, the number of separation stages required would also be very large..."(6)

> "For elimination of releases, complete recycle, isotopic removal, or a combination of these would be necessary. Isotopic removal will be very difficult; there is no proven

> system, now operational, to accomplish the separations required. Existing or proposed processes would be arranged and cascaded in a manner to accomplish the very high throughput and large isotopic separation factors required, but it seems unlikely that an economical process will be developed to handle this job."(27)

Waste releases from a recycle/separation process would be relatively small:

> "Initial calculations indicate that isotopic distillation of overhead from evaporated high-activity waste (HAW) would concentrate 95% of the tritium from processing one ton of uranium for storage in 5 to 6 gallons..."(26)

B-III.4.6 Waste Disposal

Since the above processes have tritiated water as their end-product, disposal of this waste water would not be difficult conceptually. Solidification as cement appears to be the simplest and cheapest, although even cheaper methods would perhaps be found in disposal in stagnant aquifers.

Although ERDA 76-43 mentions a wide variety of possible techniques for long-term tritium disposal, the existence of at least one cheap, simple method obviates the need for detailed discussion of more exotic methods.

B-III.4.7 Conclusions

Tritium control technology is still at the laboratory/hot-cell test stage and will not be commercially developed for many years. Sufficient proof-of-principle has occurred to give some hope that tritium control is at least possible, although the economics of this technology are highly uncertain. There is great need for detailed economic optimization studies to single out the one or two most promising systems, followed by detailed cost studies to demonstrate whether tritium control is economically feasible. Particularly interesting would be a study of recycle/separation optimization to demonstrate whether such a scheme - which is conceptually appealing - would in fact be economic and if so, at what recycle and separation rates.

B-III.5 RUTHENIUM

The following discussion of ruthenium control is abstracted from ERDA 76-43.(1)

During high-temperature solidification of waste from a fuel reprocessing facility, ruthenium may volatilize and require removal from the off-gas system. According to ERDA 76-43 the reprocessing of 1 MTU of fuel irradiated to 25,000 MWD/MTU would feed 2.5×10^5 Ci of Ru-106 into the solidification facility (based on one-year cooled waste). However, the quantities of ruthenium passing into process off-gases during waste solidification by the different types of processes are not well understood. The Final Safety Analysis Report for the AGNS Separations Facility lists the expected stack release for total Ru-106 as 4.6 millicuries per day (mCi/day); this release is expected to meet all standards and specifications promulgated by state and Federal agencies.(28)

The only data available on the release of volatile ruthenium during calcination are from laboratory measurements on simulated LWR wastes during fluid bed calcination. The mode of heating affects the quantity of gaseous ruthenium generated and passed into the off-gas system. Studies at INEL, using a simulated LWR commercial waste, show that of the ruthenium fed into a fluidized bed calciner with indirect heating, the amount leaving in the gaseous state varies from 6×10^{-4}% at 570°C to 3×10^{-2}% at 500°C, and 100% at temperatures below 375°C. When in-bed combustion heating is used, the same volatility, 2×10^{-3}%, is observed at 500°C and at 550°C.

Equipment components that can be considered for removal of both particulate and gaseous ruthenium include cyclones, quench tanks, venturi scrubbers, adsorbers and HEPA filters. Of these, the most effective in removing volatilized ruthenium appears to be the adsorber. Of a number of adsorber materials studied, silica gel (Davidson Grade 40) was found to be the most suitable. It has a high DF for volatilized ruthenium (1000), has a high capacity (exceeds 5 g Ru/liter), adsorbs at a moderate temperature, is not attacked by the off-gas, and can easily be regenerated.

The treatment of off-gases that contain volatilized ruthenium from other waste solidification processes is essentially the same as for the fluidized-bed calcination process, but few significant data are available on ruthenium behavior during solidification of LWR wastes by processes other than fluidized-bed calcination. Any differences will be directly related to the quantity of ruthenium volatilized. Some processes may release large quantities of ruthenium, which require more elaborate off-gas cleanup. Preliminary laboratory data, for example, indicate that rotary-kiln calcination might volatilize ruthenium in quantities that would require DF and adsorber capacity requirements of 500 to 5000 times those needed for fluidized-bed calcination.(1) For such a situation, a method must also be provided to prevent ruthenium plateout on pipe and component walls, which could lead to plugging. This might be done by maintaining the wall temperature between 150°C and 300°C.

An alternative to removing ruthenium from the off-gas is to use additives for holding ruthenium in the solid form. For high pH systems at high temperatures, for example, formation of aluminosilicate in the melt immobilized the ruthenium.

APPENDIX B-III

REFERENCES

1. Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle. ERDA 76-43, U.S. Energy Research and Development Administration, May 1976.

2. Allied General Nuclear Services, Barnwell Nuclear Fuel Plant Safety Analysis Report, Docket 50-332 (Rev. 3/21/64).

2.a. Ibid (Rev. 3/21/69).

3. U.S. Code of Federal Regulations, Title 40, Part 190.

4. Bendixsen, C.L. and F.O. German. 1974 Operation of the ICPP Rare Gas Recovery Facility. ICP-1057, Idaho Chemical Programs, Allied Chemical Corp., March 1975.

5. Stephenson, M.J. and R.J. Eby. Development of the 'FASTER' Process for Removing Krypton-85, Carbon-14, and Other Contaminants from the Off-Gas of Fuel Reprocessing Plants. Presented at 14th ERDA Air Cleaning Conference, August 1976.

6. Murbach, E.W., W.H. Carr and S.H. Gray. Fission Product Gas Retention Process and Equipment Design Study, ORNL-TM-4560, Oak Ridge National Laboratory, Oak Ridge, Tennessee, May 1974.

7. Foster, B.A. and D.T. Pence. An Evaluation of High Pressure Steel Cylinders for Fission Product Noble Gas Storage. ICP-1044, Idaho Chemical Programs, Allied Chemical Corp., 1974.

8. Proceedings of the International Symposium on the Management of Wastes from the LWR Fuel Cycle. CONF-76-0701, U.S. Energy Research and Development Administration, July 1976.

9. Foster, B.A., D.T. Pence and B.A. Staples. Long Term Storage Techniques for ^{85}Kr. In: Proceedings of the Thirteenth AEC Air Cleaning Conference, San Francisco, Ca., 1974. Vol. 1, U.S. Energy Research and Development Administration, CONF-740807, March 1975.

APPENDIX B-III (continued)

REFERENCES

10. Pence, D.T. and B.A. Staples. Solid Adsorbents for Collection and Storage of Iodine-129 from Reprocessing Plants. In: Proceedings of the Thirteenth AEC Air Cleaning Conference, San Francisco, Ca. CONF-740807 U.S. Atomic Energy Commission, August 1974.

11. Thomas, T.R. Separation and Retention of Iodine. In: Proceedings of the International Symposium om the Management of Wastes from the LWR Fuel Cycle, Denver, July 1976.

12. Thompson, C.H., J.A. Kelley. Evaluation of Methods for Retention of Radioiodine During Processing of Irradiated 237 Np. DP-1373, E.I. duPont deNemours & Co., Savannah River Laboratory, S.C., June 1975.

13. Groenier, W.S. and B.A. Hannaford. An Engineering Evaluation of the Iodox Process; Part 2: Removal of Iodine from Air Using a Nitric Acid Scrub in a Bubble-Cap Column; Part 3: Correlation of Mass Transfer Data; Part 4: Process Sensitivity to Impurities. ORNL-RM-4701, Oak Ridge National Laboratory, Oak Ridge, TN, May 1975.

14. Mailen, J.C. and D.E. Horner. Removal of Radioiodine from Gas Streams by Electrolytic Scrubbing. Nuclear Technology, September 1976.

15. Magno, P.J., C.B. Nelson, and W.H. Ellett. A Consideracion of the Significance of Carbon-14 Discharges from the Nuclear Power Industry. In: Proceedings of the Thirteenth AEC Air Cleaning Conference, San Francisco, Ca., CONF-740807, March 1975.

16. Public Health Considerations of Carbon-14 Discharges from the Light-Water-Cooled Nuclear Power Reactor Industry. Technical Note ORP/TAD-76-3, U.S. Environmental Protection Agency, July 1976.

17. Renfrew, C., Carbon-14 and the Prehistory of Europe. Scientific American 225(4): 63-72, October 1971.

18. Yeates, D.B., A.S. Goldin, and D.W. Moeller. Natural Radiation in the Urban Environment. Nuclear Safety 13(4): 275-286, July and August 1972.

19. Kunz, C.O. ^{14}C Gaseous Effluents from BWRs, American Nuclear Society Annual Meeting, June 8, 1975.

APPENDIX B-III (continued)

REFERENCES

20. Kunz, C.O. ^{14}C Gaseous Effluents from PWRs, Health Physics Soc., Knoxville, Tennessee, October 12, 1975.

21. Bonka, H., K. Brüssermann, G. Schwarz. Umweltbelastung durch Radiokohlenstoff aus Kerntechnischen Anlagen. Reaktortagung, Berlin, April 1974.

22. Hayes, D.W. and K.W. MacMurdo. Carbon-14 Production by the Nuclear Industry·Savannah River Laboratory, Aiken, S.C. DP-MS-74-33, to be published.

23. Final Environmental Statement--Liquid Metal Fast Breeder Reactor Program. ERDA-1535, U.S. Energy Research and Development Administration, December 1975.

24. Kelly, G.N. et al. The Predicted Radiation Exposure of the Population of the European Community Resulting from Discharge of Krypton-85, Tritium, Carbon-14, and Iodine-129 from the Nuclear Power Industry to the Year 2000. Commission of the European Communities, Luxembourg, Doc.V/2676/75, September 1975

25. Ribnikar, S.V. and J.D. Pupezin. Possibilities of Tritium Removal from Waste Waters of Pressurized Water Reactors and Fuel Reprocessing Plants. In: Proceedings of the Thirteenth AEC Air Cleaning Conference, CONF-740807, San Francisco, California, August 1974.

26. Savannah River Laboratory Quarterly Report. Light Water Reactor Fuel Recycle. DPST-LWR-76-1-2, E.I. Dupont de Nemours & Co., April-June 1976.

27. Musgrave, B.C. Tritium Distribution in the Nuclear Industry. ICP-1041, Allied Chemical Corporation, Idaho Falls, Idaho, January 1974.

28. Barnwell Nuclear Fuel Plant Separations Facility Final Safety Analysis Report. Allied General Nuclear Services, 1974.

29. Hall, N.E. and G.N. Ward. Tritium Control by Water Recycle in a Nuclear Fuel Reprocessing Plant. NEDG-11342, General Electric Company, San Jose, California, June 1975.

APPENDIX B- III (continued)

REFERENCES

30. Goode, J.H. Voloxidation, Removal of Volatile Fission Products from Spent LMFBR Fuels. ORNL-TM-3723, Oak Ridge National Laboratory, Oak Ridge, Tennessee, 1973.

31. Unger, W.E. LMFBR Fuel Cycle Studies Progress Report for November 1970. ORNL-TM-3250 No. 21 Oak Ridge National Laboratory, Oak Ridge, Tennessee, 1970.

32. Stevens, W.H. and F.P. Blackstein. Heavy Water Production Using Hydrogen-Water Exchange. AECL Research and Development in Eng., Winter 1973, Atomic Energy of Canada, Ltd., AECL-4606, Winter 1973.

APPENDIX B-IV

DETAILS OF REFERENCE DISPOSAL-SITE FACILITIES

This appendix describes details of the reference disposal site discussed in Section 3.5.1 of this report.

B-IV.1 SURFACE FACILITIES

B-IV.1.1 Canned Waste Receiving Building

This building must receive approximately 350,000 canisters of high-level waste (HLW), cladding hulls, and intermediate-level TRU waste over the life of the facility, and must be sized accordingly. For a ten-year operating life this corresponds to a throughput of approximately 100 canisters (8-10 rail transportation casks) per day for a seven-day-per-week operation. The canister receiving building must provide cask-handling capability -- i.e., for rail car cask unloading, cask decontamination as necessary, cask cool-down, and cask repair. An optimized HLW cask, which is shipped in a vertical orientation and is capable of being unloaded without removal of the cask from the rail car, will reduce cask-handling time. It is anticipated that average rail cask turnaround time will be on the order of 12-24 hours. Alternate forms of packaging for intermediate-level TRU, such as shielded bulk shipment, will greatly reduce the service demand on the canister-receiving facility.

B-IV.1.1.1 Surface Canister-Handling Cells

Canister-handling cells must be designed to handle the canister throughput discussed above. The canister-handling cells must have the capability to remove, temporarily store, and replace the shipping cask head; unload canisters from the shipping cask; inspect individual canisters for damage; repair or package damaged canisters; and transfer acceptable canisters either to the canned waste shaft hoist cell for subsequent transfer to the mine or to the interim surface-storage facility. All canister handling must be done remotely because of the high radiation levels of the canisters. The in-cell equipment must be specifically designed for remote maintenance, or the various cells must be segregated in order that a cell with faulty equipment may be isolated, and decontaminated if necessary, to give personnel access for hands-on maintenance. Because of the sealed design of the canisters, relatively low contamination and radiation levels are anticipated in the cells when waste canisters are not being handled.

B-IV.1.1.2 Interim Waste Canister Storage

An interim canister-storage facility should be constructed to accept an overflow of canisters if there are problems in the canister hoist system or in the mine. Canisters would be separated according to waste type and placed in dry storage vaults. Cooling would be provided either by a network of cooling coils distributed throughout the storage vault or by forced air circulation around the canisters. Final heat removal from the cooling system would be via a heat exchanger for either option. An interim storage capability of 3-6 months of normal canister receipt operations has been suggested.

B-IV.1.1.3 Canister Hoist Cell/Hoist Equipment

Canisters are transferred from the canister-handling cells to the canister-hoist cell and loaded one at a time into the canister hoist cage for transfer to the mine. High-speed hoisting equipment will be located outside the cell to permit hands-on maintenance.

B-IV.1.2 Low-Level TRU Receiving Building

This facility must be designed to receive approximately 140,000 m^3 over the life of the facility. The receiving building should be designed to permit receipt of low-level TRU waste either by rail or truck. Most shipments will probably be made by truck with an average shipment containing approximately the equivalent of 96 208-liter (55-gallon) drums or 20 m^3 of waste. For a facility that will be filled in ten years, the average throughput will be 2.5-3 truckloads per day for a five-day-per-week operation, and truck unloading time should average approximately two hours. Trucks will be unloaded using conventional freight-handling equipment, such as fork lifts or overhead cranes. If cladding hulls and intermediate-level TRU wastes are shipped in bulk package form instead of in canisters, these containers will be received in the low-level TRU facility, with the design adjusted to handle the increased throughput. The capacity of the low-level TRU interim storage building must also be increased accordingly.

B-IV.1.2.1 Low-Level TRU Interim Storage Building

This building will be used to store temporarily any backlog of received waste that might result from problems with the hoist equipment or in the low-level TRU portion of the mine. This building will resemble an ordinary warehouse and should have a capacity of 3-6 months' accumulation of low-level TRU.

B-IV-1.2.2 Low-Level TRU Hoist Facility

This facility will contain high-speed hoisting equipment and a transfer cage loading area.

B-IV.1.3 Man and Materials (M&M) Hoist Facility

This facility will house the high-speed M&M hoisting equipment. It will also serve as a staging area for personnel and materials transfer into the mine, the fresh air inlet for the mine ventilation system, and the transfer point for the mined materials storage and reclamation system.

B-IV.1.4 Mine Ventilation System

The mine ventilation system will provide a continuous, once-through source of pre-filtered fresh air into the mine through the ventilation compartment of the M&M shaft. It will also distribute the air throughout the mine, as needed, from least-contaminated to potentially most-contaminated areas, and then exhaust the air through the ventilation exhaust shaft into the radwaste treatment system. The ventilation system must be designed to treat all ventilation exhaust in the event of an abnormal occurrence in order to ensure the safety of the general public. A detailed discussion of a typical mine ventilation system suitable for use in the reference facility has been developed for the Office of Waste Isolation.(1) The facility ventilation system can also be used to transport waste heat out of the mine storage facilities during any period for which retrievability of wastes is desired.

V-IV.1.5 Site Radwaste Process Facility

The facility will process, package, and dispose of all radioactive wastes generated at the waste burial site. All potentially contaminated exhaust gases from the various on-site waste-handling cells, the chemical laboratory, and the mine ventilation system will be passed through a series of filters in the gaseous radwaste treatment system prior to release from the plant stack. The radioactivity released from the stack will be constantly monitored to ensure that it meets appropriate regulatory guidelines. Liquid radwaste, mostly in the form of laboratory wastes, decontamination solvents, and laundry wastes will be processed into a solid waste form for burial. Solid wastes will include contaminated tools and equipment, miscellaneous health physics and laboratory supplies, solidified liquid radwaste, spent filters from the gaseous radwaste treatment system,radiation-protective clothing that can no longer be used, etc. Solid radwaste will be suitably packaged for ultimate burial in the LLW portion of the mine facility.

B-IV.1.6 Mined Storage and Reclamation System

For the non-retrievable reference facilities in this study, a minimum of $0.9 \times 10^6\ \mathrm{m}^3$ ($1.2 \times 10^6\ \mathrm{yd}^3$) of salt for an HLW facility and $1.8 \times 10^6\ \mathrm{m}^3$ ($2.3 \times 10^6\ \mathrm{yd}^3$) of salt for a throwaway-fuel-cycle facility must be removed from the mine during construction, stored for the duration of the facility operating period, and then returned to the mine as backfill during facility decommissioning. For a fully-retrievable facility, such as the one discussed at the end of this section, the corresponding volumes are $14 \times 10^6\ \mathrm{m}^3$ ($18 \times 10^6\ \mathrm{yd}^3$) for the HLW and $26 \times 10^6\ \mathrm{m}^3$ ($34 \times 10^6\ \mathrm{yd}^3$) for spent fuel. The mined materials storage and reclamation system will consist of: 1) a rail loop transportation system connecting the M&M shaft with the designated mined materials storage area; 2) hopper-type rail cars to transport the mined salt; and 3) salt-handling equipment for rapid loading of hopper cars at the M&M shaft and piling of the unloaded salt at the designated storage location. The stored salt should be protected from the weather during the storage period to prevent salt contamination of the environment.

B-IV.1.7 Site Support Facilities

These facilities include an administration building, machine shop, warehouse, personnel staging area -- including lunch room, locker room, and showers --, health physics and chemical laboratory facilities, and a site security system including perimeter fences, gate houses, etc.

B-IV.1.8 Shaft Data

Four concrete-lined shafts will be required for the reference facilities assumed in this study. The following shaft dimensions have been suggested:(1)

Man and Materials Shaft -- 7.9 m (26 ft) i.d.
Canister Waste Shaft -- 1.8 m (6 ft) i.d.
Low-Level TRU Shaft -- 3.0 m (10 ft) i.d.
Ventilation Exhaust Shaft -- 6.1 m (20 ft) i.d.

A typical shaft-digging sequence is detailed, and suitable operating criteria for hoisting equipment are outlined.(1)

B-IV.2 MINE COMPONENTS

The design developed for the Office of Waste Isolation is for retrievable storage.(1) To increase mine stability while minimizing overall mine volume, the TRU fields have been located 21 m (70 ft) above the HLW burial field.

B-IV.2.1 Receiving Stations

B-IV.2.1.1 M&M Receiving Station

This station is located at the bottom of the M&M shaft. All personnel, materials, and equipment entering and leaving the mine do so through this station. This station serves the following functions:

- Provides a salt transfer point to the surface during mine excavation;
- Serves as mine operations control, including a personnel staging area, offices, toilets, communications system, etc.;
- Controls ventilation by means of intake air distribution louvers;
- Handles all materials entering the mine, including many large and heavy objects, such as disassembled parts of the canned waste placement vehicles. This station must have the handling equipment and facilities to receive and assemble such components;
- Connects, through the elevator system, the main (HLW) level of the mine with the TRU storage areas 21 m (70 ft) above the main mine level.

B-IV.2.1.2 High Level Waste Receiving Station/Transfer Cell

This facility is a remote-handling cell at the base of the canned waste hoist shaft. Equipment in the cell must be capable of receiving canned waste from the surface, temporarily storing canisters (10-15 maximum) prior to burial, and transferring canisters to the waste placement vehicle. Waste canisters will be loaded into the waste placement vehicle through ports in the top of the transfer cell. The number of loading ports is determined by the required waste placement rate. Each waste placement vehicle will be capable of handling an average of three canisters per normal eight-hour shift, and one port can efficiently service three vehicles.

B-IV.2.1.3 Intermediate-Level TRU Waste Receiving Station/Transfer Cell

This remote canister-handling cell is located at the intermediate-level TRU storage level of the mine, approximately 21 m (70 ft) above the HLW receiving station. The canned waste shaft passes through the TRU transfer cell; canned intermediate-level TRU is removed with remotely operated equipment at the intermediate-level TRU receiving station, while canned HLW continues down the waste shaft to the HLW receiving facility. (Facilities with separate HLW and TRU shafts are also being considered.) The TRU transfer cell must provide the same functions for intermediate-level TRU canisters as the HLW transfer cell does for HLW.

B-IV.2.1.4 Low-Level TRU Waste Receiving Station

This is a modest transfer area located at the base of the low-level TRU shaft. It must be large enough to permit efficient handling of containers with a fork lift vehicle.

B-IV.2.2 Mine Level Machine Shop/Equipment Service Area

An area is set aside in the mine to service the waste placement equipment. This area should also include a machine shop in order that routine repairs may be made in the mine rather than requiring the time-consuming transfer of equipment to a surface facility. The service area should be connected by a series of corridors to the HLW and TRU, storage areas in the same service facility in order to make it possible to service equipment from these areas.

B-IV.2.3 HLW Storage Area

The storage area for the reference HLW facility is made up of 1112 burial tunnels. Adjacent tunnels are parallel to each other with 23.8-m (78-ft) center-to-center spacing, and are connected by a series of main and branch access corridors. Each burial tunnel is essentially identical in design, measuring approximately 171 m (560 ft) long x 5.5 m (18 ft) wide x 5.5 m (18 ft) high. The height and width dimensions are set by spatial requirements of handling equipment and may have to be modified slightly when a final design is evolved for a canned waste placement vehicle. Thirty-two canister holes are bored in the tunnel floor on 5.3-m (17.5-ft) center-to-center spacing. Each tunnel has only one entrance, and each exhausts to the ventilation treatment system from the dead end of the tunnel. This arrangement makes it possible always to circulate air from a less-contaminated to a potentially more-contaminated area of the mine. Canisters are transported to the burial tunnel with the canister placement vehicle discussed later and are placed in canister holes, working from the dead end of the tunnel toward the open end. After the canister has been lowered into the burial hole, the hole is backfilled with crushed salt. When all canister holes in the burial tunnel are full, the tunnel is backfilled as soon as convenient with salt removed from a burial tunnel under construction.

Two rows of burial tunnels are located back-to-back (dead end to dead end) in order to minimize the number of branch connecting tunnels and the amount of excavation required for the network of ventilation exhaust tunnels around the HLW storage area.

Main and branch corridors lead from the HLW receiving-transfer cell to the HLW burial field. Approximate dimensions are 9.1 m (30 ft) wide and 5.5 m (18 ft) high, with actual dimensions set by the requirements of the handling equipment.

B-IV.2.4 Canister Holes

Each burial tunnel will have 32 canister holes bored along the centerline. HLW canister holes will be 0.46 m (18 in) in diameter x 6.1 m (20 ft) long. After a canister is in place, the remainder of the hole will be backfilled with crushed salt to provide radiation shielding. The crushed salt will be put in with the canister transport still in place over the canister hole to provide shielding for the operators. Once the hole is backfilled, the dose rate at the floor of the burial tunnel will be less than 10 mR/hr. Spent-fuel canister holes will contain up to four canisters per hole. Each hole will be 0.46 m (18 in) in diameter and 23.2 m (76 ft) deep. Prior to loading, each canister hole will be fitted with a shielded isolation valve discussed in the following section. The loading sequence will be as follows:

(1) Position the canister transport vehicle over the shielded isolation valve.

(2) Open the isolation valve and lower waste canister No. 1 into the empty canister hole and release canister.

(3) Backfill hole with a premeasured volume of crushed salt to approximately 0.6 m (2 ft) above the in-place canister. This backfill of salt will provide a floor on which the next canister can rest.

(4) Close isolation valve and return to the waste receiving/transfer cell for canister No. 2.

(5) Repeat steps 1 through 4 for canisters Nos. 2, 3, and 4.

(6) After placement of canister No. 4, backfill to the top of the canister hole.

The multiple loading of four spent-fuel canisters in one hole produces less heat per location than a single HLW canister. The alternative to multiple loading of spent-fuel canisters is boring approximately four (actually 3.6) times as many holes in each standard burial tunnel. Increasing the number of canister rows in the tunnel would require widening the tunnel, thereby increasing overall facility costs.

B-IV.2.5 Canned Waste Placement Vehicle

Canned wastes, including HLW, spent fuel, cladding hulls, and intermediate-level TRU are highly radioactive and must be transported in specially designed, heavily-shielded vehicles. Each vehicle must provide the following functions:

- Provide a fine positioning capability for the transport vehicle in centering the transport vehicle cask opening over the transfer cell port and the canister burial holes.

- Provide shielding to operating personnel during the canister loading/unloading sequences through the use of a movable shield. Shielding must be effective against both neutron and gamma radiation.

- Provide internal, remotely-connected capability for moving canisters into the transport vehicle from the transfer cell and for lowering canisters into the canister burial holes. For spent fuel and TRU the hoisting/lowering capability may be as much as 24 m (80 ft).

- Provide motive power for waste transfer. The most probable power alternatives are diesel and electric. Round-trip distances per canister may be as great as 4 miles for HLW and 8 miles for spent fuel.

- Provide shielding and, if necessary, cooling capability during transport. Shielding must be effective against both neutron and gamma radiation.

- Provide capability for remote viewing of the canned waste handling activity through the use of a radiation-tolerant television camera system.

- Provide crushed salt backfill capability.

- Provide constant, portable radiation detection capability.

The above functions may be provided by one or several pieces of equipment. The transfer vehicle will probably resemble a spent-fuel shipping cask in a vertical position mounted on a motorized bed set on either rubber tires or tracks. An internal hoist located in the top of the cask portion of the vehicle would raise and lower the canister. A thick shielded gate valve would be located at the bottom of the cask to provide shielding from radiation coming through the opening in the bottom of the cask during canister transport. The transport vehicle would be 5 m (16 ft) high and weigh 30-45 MT, for an HLW facility. The length and width dimensions will depend on the ultimate equipment design, mode of propulsion, etc. but are expected to be no more than 4.6 m (15 ft) long by 3.0 m (10 ft) wide. The transport vehicle for spent fuel will be 6 m (21 ft) high and weigh 35-55 MT. The base dimensions will be similar to the HLW machine. The interface between the transfer cell or the canister hole and the transport vehicle can be accomplished either by: 1) using a separate shielded isolation valve; 2) lowering a shielded collar suspended from the bottom of the transport vehicle; or 3) lowering the cask itself until it touches the floor. An isolation valve of some sort will be necessary to close the port over the transfer cell when the transporter is not in place for loading. Such a valve will also be necessary if multiple canisters are to be loaded into spent-fuel and TRU canister holes. Furthermore, an isolation valve must be developed to facilitate canister-hole shield-plug installation if retrievable storage is to be used. It is probable that a combination of an isolation valve and a shield-collar or lowered-cask technique will be used to provide personnel shielding during canister transfer.

Canister transport vehicle work has been done for Project Salt Vault.(2) Moreover, the functional requirements for canister handling are very similar to those encountered in High-Temperature Gas-cooled Reactor (HTGR) refueling experience.

B-IV.2.6 Intermediate-Level TRU Waste Storage Field

The intermediate-level TRU facility layout in Reference 1 has been assumed for this study. Canned cladding hulls and TRU are received at the intermediate-level TRU receiving station/transfer cell, loaded into canned waste transporter vehicles, transferred to the TRU canister storage field, and lowered into canister holes. The procedure is identical to that for HLW disposal. Burial tunnels are 171 m (560 ft) long x 11.6 m (38 ft) wide x 5.5 m (18 ft) high, and contain six parallel rows of canister holes. Canisters are located on a square array with 1.2-m (4-ft) center-to-center spacing. Each burial tunnel can store 790 canisters; 398 rooms are required to accommodate the 313,500 canisters to be buried in the intermediate-level TRU field. There are salt pillars 12.2 m (40 ft) thick between adjacent rooms, aligned in parallel on 23.8-m (78-ft) center-to-center spacing. When all canister holes in a room are filled, the burial tunnel is backfilled with salt being removed from the construction of new tunnels.

The reference arrangement described above may not be the best conceivable design. Because TRU waste produces relatively low levels of decay heat, multiple canister loading per hole or stacked warehouse-type storage of shielded bulk shipping containers are both possible. Both approaches should be investigated before deciding on a final TRU canister storage field design.

B-IV.2.7 Low-Level TRU Waste Burial Field

Low-level TRU gives off low levels of both heat and radiation and can therefore be handled with conventional freight-handling equipment. TRU drums are picked up at the low-level TRU receiving station with a diesel or electric powered fork lift vehicle and transported to burial tunnels where units are stacked, warehouse-style. Each burial tunnel measures 171 m (560 ft) long x 11.6 m (38 ft) wide x 5.5 m (18 ft) high. When a burial tunnel is filled to capacity, it is backfilled with salt from tunnels under construction in another part of the mine.

The above array may not represent the best storage for TRU drums. The size of the low-level TRU storage field can be decreased by placing some (possibly all) of this waste in HLW burial tunnels before they are backfilled with salt.

REFERENCES

1. Parsons, Brinckerhoff, Quade & Douglas, Inc. Waste Isolation Facility Description - Bedded Salt. Document Y/OWI/SUB-76/16506, September 1976.

2. Bradshaw, R. and W. McClain, Project Salt Vault: A Demonstration of the Disposal of High-Activity Solidified Wastes and Underground Salt Mines. ORNL-4555, Oak Ridge National Laboratory, April 1971.

APPENDIX B-V

CORROSION OF METAL CONTAINERS

B-V.1 TEMPORARY STORAGE OF CONTAINERS IN WATER

If the container is temporarily stored in water prior to burial, it must resist corrosion. The water will be deionized; its chemistry will have to be monitored and it will have to be kept free of chlorides that could enhance corrosion of some container materials, such as stainless steel. There is also the remote possibility that corrosion could be enhanced by the products formed by radiolysis of water (oxygen and peroxides). A report from Sweden mentions the radiolysis effect on corrosion.(1)

The container corrosion problem has been discussed with personnel at Battelle Northwest Laboratories, which is planning to make full-sized containers of various metals, including carbon steel, stainless steel, and Inconel, and expose them to water. Chlorides will be introduced as incursions to the water in a worst-case situation. Possible harmful effects resulting from sensitization of the stainless steel or from radiolysis of the water will be found from these tests. Radiolysis products of water are not believed to constitute a problem, except if there are crevices in the metal.

B-V.2 LONG-TERM STORAGE IN SALT BRINE ENVIRONMENT

Data have been compiled on corrosion rates of alloys that may be used as container materials and are given in Tables B-V-1 through B-V-5. If the container is exposed to hot brine in a salt deposit, the alloys having high resistance to general corrosion or pitting corrosion are titanium-nickel (Ti Code-12) or titanium-palladium alloys. If the container is subjected to hot salt above the boiling point of water in an atmosphere containing water and oxygen, the alloy may be subjected to pitting attack. This attack may occur with long-time exposure at temperatures above several hundred degrees Celsius. The mechanism of attack has been described in a recent paper.(2) Hot salt pitting has been discussed with several experts on corrosion at Titanium Fabrication Corporation (titanium alloys), and at the International Nickel Company (stainless steels and nickel-based alloys). These discussions indicated that titanium alloys may not be resistant to pitting in hot salt, but there is no information available on pitting times. Inconel 625 or Hastelloy C would be the alloys most resistant to pitting in hot salt, but information on hot salt pitting of these alloys below 300°C is also not available. Given the lack of information, it is recommended that tests be carried out to determine the pitting resistance of selected alloys, such as Inconel 625 or Hastelloy C, in hot salt.

In Tables B-V-1 through B-V-5 some corrosion rates are given as 0. This simply means that the corrosion was below the detectable limit for the experiment in question. To gain an idea of the limit of detectability, it is suggested that in each case the reader look at the reference number in the last column of each table, and scan the tables for other corrosion rates reported by the same reference. If no such data are found,, reference must be made to the original paper.

TABLE B-V-1

CORROSION OF TITANIUM ALLOYS IN SODIUM CHLORIDE SOLUTIONS

Alloy	Corrodent	Temperature	Time on Test	Corrosion Rate, mm/yr	Corrosion Rate, mils/yr	Pitting-Crevice Corrosion	Reference No.
Ti	3% NaCl	Boiling	--	0.0003	0.01	--	3
Ti	29% NaCl	110°C (230°F) (Autoclave)	--	0.003	0.1	--	3
Ti	Sat. NaCl	Boiling 107°C (225°F)	--	0.001	0.05	--	3
Ti	Sea water, natural	Ambient	4.5 yrs.	0.0008	0.03	None in >5 yrs	4
Ti-6Al -4V	Sea water, natural 0 velocity	Ambient	16 weeks	<.003	<0.1	--	5
Ti-6Al -4V	Sea water, natural, 5 m/sec. velocity	Ambient	2 weeks	0.0038	0.15	--	5
Ti	Conc. Sea water, 1.04 - 1.63x	24-41°C (75-105°F)	134 days	$<3\times10^{-5}$	<0.001	--	6
Ti-.5Pd	Conc. Sea water, 1.04 - 1.63x	24-41°C (75-105°F)	134 days	$<3\times10^{-5}$	<0.001	--	6
Ti	Buried in Tidal Marsh	Ambient	7.8 yrs.	0.	0.	No pits	7
Ti-Ni (.1-5%)	Sea water, synthetic	200-288°C (392-550°F)	504 hrs.	0.	0.	None	8
Ti	Sea water, synthetic	200°C (392°F)	96 hrs.	--	--	Crevice corrosion under Teflon gasket	8
Ti	25% NaCl	Boiling	48 hrs.	0.	0.	--	9

TABLE B-V-1
(Continued)

Alloy	Corrodent	Temperature	Time on Test	Corrosion Rate,		Pitting-Crevice Corrosion	Reference No.
				mm/yr	mils/yr		
Ti-.2Pd	25% NaCl	Boiling	48 hrs.	0.	0.	--	9
Ti	Sat. NaCl	Ambient	--	0.	0.	--	10
Ti-.15Pd	NaCl Brine	93°C (200°F)	--	1.5×10^{-5}	0.0006	--	10
Ti	23% NaCl in contact with Teflon	Boiling	--	--	--	Crevice attack	10
Ti-50A	Sea water, natural	2-121°C (35-250°F)	--	--	--	No pitting	11
Ti-50A	25% NaCl	2-82°C (35-180°F)	--	--	--	No pitting	11
Ti-50A	25% NaCl	82-149°C (180-300°F)	--	--	--	Pits possible in metal-to-metal crevices	11
Ti-50A	25% NaCl	Above 149°C (300°F)	--	--	--	Pits	11
Ti	23% NaCl pH 1.4-1.5	Boiling	7 days	0.	0.	--	12
Ti	23% NaCl pH 1.2-1.3	Boiling	7 days	0.64-0.71	25-28	--	12
Ti	Sea water, 1.5x conc.	116°C (240°F)	6 yrs.	No corrosion failures in 90 miles of 22-gauge tubing			13
Ti-2Ni	3.5% NaCl, pH = 1.0	Boiling	500 hrs.	No stress corrosion cracking at 90% of 0.2% offset yield			14
Ti	1M NaCl pH = 3.0	149°C (300°F)	246 hrs.	--	--	Extensive crevice corrosion	18

TABLE B-V-1
(Continued)

Alloy	Corrodent	Temperature	Time on Test	Corrosion Rate, mm/yr	Corrosion Rate, Mils/yr	Pitting-Crevice Corrosion	Reference No.
Ti	Sat. NaCl pH 7.5	107°C (225°F)	--	0.0036	0.14	Sl. crevice attack	19
Ti	Sat. NaCl pH >12	98°C (208°F)	--	0.0036	0.14	None	19

Source: Based on references listed above.

TABLE B-V-2

CORROSION OF INCONEL ALLOYS IN SODIUM CHLORIDE SOLUTIONS

Alloy	Corrodent	Temperature	Time on Test	Corrosion Rate, mm/yr	mils/yr	Pitting-Crevice Corrosion	Reference No.
625	Sea water, natural 0 velocity	Ambient	16 wks.	<.003	<0.1	--	5
625	Sea water, natural 5 m/sec. velocity	Ambient	2 wks.	8×10^{-4}	0.03	--	--
625	Sea water, quiet	Ambient	7 yrs.	<.003	<0.1	Incipient crevice beneath fouling	15
625	Sea water, 0.6 m/sec	Ambient	5.9 yrs.	<.003	<0.1	0.28 mm (11 mils) beneath Teflon	15
825	Sea water, quiet	Ambient	3 yrs.	0.003	0.1	.0002 mm (.007 mils) max. under barnacles	15
Welded or Plain 718	Sea water, quiet	Ambient	6 mo.	0.003	0.1	--	16
600	Sea water	Ambient	2.7 yrs.	0.002	0.08	Max. pitting--1.6 mm (63 mils)	17
600	Saturated Salt Brine pH = 7.5	107°C (225°F)	185 days	0.027	1.05	Pitting	19
600	Saturated Salt Brine pH >12	98°C (208°F)	125 days	0.134	5.27	Severe pitting and crevice corrosion	19

Source: Based on references listed above.

TABLE B-V-3

CORROSION OF STAINLESS STEELS IN SODIUM CHLORIDE SOLUTIONS

Alloy	Corrodent	Temerature	Time on Test	Corrosion Rate,		Pitting-Crevice Corrosion	Reference No.
				mm/yr	mils/yr		
304	Sea water, fouling	Ambient	160 days	0.038	1.5	Pits perforated samples	3
316	Sea water, fouling	Ambient	160 days	<.003	<0.1	--	3
304	14% NaCl + 12% KCl	25°C (77°F)	38 days	0.008	0.3	Pits .03-.13 mm (1-5 mils) in crevice	3
316	14% NaCl + 12% KCl	25°C (77°F)	38 days	0.008	0.3	Pits .03-.13 mm (1-5 mils) in crevice	3
304	Conc. sea water, 1.04-1.63x partly deaerated	24-41°C (75-105°F)	134 days	0.0002	0.008	--	6
316	Conc. sea water, 1.04-1.63x partly deaerated	24-41°C (75-105°F)	134 days	0.0002	0.009	--	6
304	Sea water, quiet	Ambient	3 yrs.	--	--	max. pitting 3.0 mm (119 mils), avg. pitting 1.1 mm (42 mils)	15
316	Sea water, quiet	Ambient	3 yrs.	--	--	max. pits 1.8 mm (72 mils), avg. pits 1.3 mm (50 mils)	15
304	4% NaCl aerated pH 7.0	90°C (194°F)	--	0.15	5.8	av. max. pit depth = .56 mm (22 mils) 62 pits/sq dm	17
316	4% NaCl, aerated	90°C (194°F)	24 hrs.	0.01	0.5	No pits	17

TABLE B-V-3
(Continued)

Alloy	Corrodent	Temperature	Time on Test	Corrosion Rate,		Pitting-Crevice Corrosion	Reference No.
				mm/yr	mils/yr		
304	Sea water	Ambient	1 yr.	0.008-0.01	.3-.5	Pitted	17
316	Sea water	Ambient	1 yr.	0.0001	0.002	No pits	17
316	Saturated salt brine pH >.5	107°C (225°F)	185 days	0.01	0.5	Stress and crevice corrosion	19
316	Saturated salt brine pH >12	98°C (208°F)	125 days	0.97	38	Stress cracking. Crevice corrosion	19
310	Sea water	Ambient	42 months	0.008	0.3	Pit depths--max. 4.1 mm (163 mils), avg. 1.8 mm (70 mils)	20
316	Sea water	Ambient	42 months	0.005	0.2	Pit depths--max. 1.6 mm (62 mils), avg. 0.8 mm (32 mils)	20

Source: Based on references listed above.

TABLE B-V-4

CORROSION OF MILD STEEL IN SODIUM CHLORIDE SOLUTIONS

Steel Alloy	Corrodent	Temperature	Time on Test	Corrosion Rate, mm/yr	Corrosion Rate, mils/yr	Pitting-Crevice Corrosion	Reference No.
Carbon steel	Sea water	Ambient	1 yr	0.104	4.1	—	3
2.7 Ni	Sea water	Ambient	1 yr	0.097	3.8	—	3
Mild	Synthetic sea water aerated	24-29°C (75-85°F)	3 months	0.157	6.2	—	3
Mild	Synthetic sea water aerated	60-63°C (140-145°F)	3 months	0.239	9.4	--	3
Mild	3% NaCl	Ambient	--	--	--	Relative corr. rate = 2.1	4
Mild	25% NaCl	Ambient	--	--	--	Relative corr. rate = 0.4	4
Carbon	Buried in Tidal Marsh	Ambient	7.8 yr	0.198	7.8	Max. pit depth = 3.3 mm (128 mils)	7
Carbon	Sea water	Ambient	15 yrs	0.08-.12	3.0-4.8	Max. pit depth = 1.1-6.1 mm (43-240 mils)	17
Carbon	Sea water	Ambient	42 months	1.60	6.3	Max. pit depth 2.1 mm (81 mils) avg. 1.5 mm (61 mils)	20
Mild	Vel. = 1.5 m/sec; O_2= 1 ppm Sea water, 83% salinity	82°C (180°F)	30 days	1.52	60	—	21

Source: Based on references listed above.

TABLE B-V-5

COMPARISON OF THE CORROSION RATE OF ZIRCONIUM WITH OTHER ALLOYS IN CHLORIDE SOLUTIONS

Alloy	Corrodent	Temperature	Time on Test	Corrosion Rate,		Pitting-Crevice Corrosion	Reference No.
				mm/yr	mils/yr		
Ti-A-55	55% $CaCl_2$ saturated	113°C (235°F)	84 days	0.	0.	--	22
Comm. Zr	55% $CaCl_2$ saturated	113°C (235°F)	84 days	0.01	0.4	--	22
Inconel	55% $CaCl_2$ saturated	113°C (235°F)	84 days	0.76	30.	--	22
Nionel	55% $CaCl_2$ saturated	113°C (235°F)	84 days	0.74	29.	--	22
Mild Steel	55% $CaCl_2$ saturated	113°C (235°F)	84 days	1.32	52.	--	22
Ti-A-55	73% $CaCl_2$	177°C (350°F)	49 days	0.56	22.	Severe pitting	22
Comm. Zr	73% $CaCl_2$	177°C (350°F)	49 days	>3.0	>118.	Sample dissolved	22
Inconel	73% $CaCl_2$	177°C (350°F)	49 days	0.08	3.	Slight pitting	22
Nionel	73% $CaCl_2$	177°C (350°F)	49 days	0.03	1.	Moderate pitting	22
Mild Steel	73% $CaCl_2$	177°C (350°F)	49 days	2.90	114.	--	22
Ti-A-55	Saturated NaCl	21°C (70°F)	208 days	0.	0.	--	22

TABLE B-V-5
(Continued)

Alloy	Corrodent	Temperature	Time on Test	Corrosion Rate, mm/yr	Corrosion Rate, mils/yr	Pitting-Crevice Corrosion	Reference No.
Comm. Zr	Saturated NaCl	21°C (70°F)	208 days	0.0001	0.004	--	22
Mild Steel	Saturated NaCl	21°C (70°F)	208 days	0.08	3.	Moderate pitting	22
Ti-A-55	Saturated NaCl	60°C (140°F)	169 days	0.0002	0.006	--	22
Comm. Zr	Saturated NaCl	60°C (140°F)	169 days	0.0002	0.007	--	22
Mild Steel	Saturated NaCl	60°C (140°F)	169 days	0.20	8.	Slight pitting	22
Ti-A-55	Saturated NaCl	71°C (160°F)	163 days	0.	0.	--	22
Comm. Zr	Saturated NaCl	71°C (160°F)	163 days	0.0003	0.01	--	22
Mild Steel	Saturated NaCl	71°C (160°F)	163 days	0.66	26.	--	22
Zr	20-25% NaCl	Boiling	--	<.008	<0.3	—	3
Zr	Sea Water	Ambient	15.5 wks	0.003	0.1	—	4

Source: Based on references listed above.

Appendix B-V

REFERENCES

1. Buried Nuclear Waste Must Be Enclosed in Gold to Resist Corrosion. Anti-Corrosion, June 1977.
2. Meyers, J.R. and J.A. Hall. Hot Salt Stress Corrosion Cracking of Titanium Alloys: An Improved Model for the Mechanism. Corrosion, 33: 7, July 1977.
3. LaQue, F.L. and H.R. Copson. Corrosion Resistance of Metal and Alloys. Reinhold Publishing Corp., N.Y., 2nd ed., 1963.
4. Uhlig, H.H.. Corrosion and Corrosion Control. John Wiley & Sons, Inc., N.Y., 1965.
5. Davis, J.A. and G.A. Gehring. The Effects of Velocity on the Sea Water Corrosion Behavior of High Performance Ship Materials. Materials Performance, 14 (4): 32-39, April 1975.
6. Schrieber, C.F. T.D. Boyce, B.D. Oakes, and F.H. Coley. Behavior of Metals in Desalting Environments--Sixth Progress Report. Materials Performance 14 (2): 9-14, February 1975.
7. Sanderson, B.T. and M. Romanoff. Performance of Commercially Pure Titanium in Corrosive Soils. Materials Protection, 8 (4): 29-32, April 1969.
8. Bomberger, H.B., and L.F. Plock. Methods Used to Improve Corrosion Resistance of Titanium. Materials Protection, 8 (6): 45-48, June 1969.
9. Greco, E.C.. The Catastrophic Corrosion Behavior of Titanium in Chemical Processes. Materials Protection, 6 (10): 22-27, October 1967.
10. Titanium Design Data Book, Titanium Metals Corp. of America, N.Y.
11. Feige, N.G. and R.L. Kane. Experience with Titanium Structures in Marine Service. Materials Protection and Performance, 9 (8): 13-16, August 1970.
12. Millaway, E.E.. Titanium: Its Corrosion Behavior and Passivation. Materials Protection, 4 (1): 17-21, January 1965.

Preceding page blank

Appendix B-V (Continued)

REFERENCES

13. Newton, E.H., J.D. Birkett, and J.M. Ketteringham. Survey of Materials Behavior in Large Desalting Plants Around the World. ADL Report to the Office of Saline Water, U.S. Department of the Interior, Washington, D.C., March 1972.

14. Sedricks, A.J., J.A.S. Green, and D.L. Novak. Electrochemical Behavior of Ti-Ni Alloys in Acidic Chloride Solutions. Corrosion, 28 (4): 142, April 1972.

15. Huntington Alloy Products Div., The International Nickel Co., Inc. Resistance to Corrosion. Huntington, W. Va., 1970.

16. Hohman, A.E. and W.L. Kennedy. Corrosion and Material Selection Problems on Hydrofoil Craft. Materials Protection, 2 (9): 59, September 1963.

17. Uhlig, H.H., Ed. Corrosion Handbook. John Wiley & Sons, Inc., N.Y., 1948.

18. Griess, J.C., Jr. Crevice Corrosion of Titanium in Aqueous Salt Solutions. Corrosion, 24 (4): 97, April 1968.

19. Turnbull, J.M. Salt Refinery Corrosion Caused by Varying Evaporation Conditions. Corrosion 16 (7): 11-16, July 1960.

20. LaQue, F.L. and A.H. Tuthill. Economic Considerations in the Selection of Materials for Marine Applications. Trans. Soc. of Naval Architects and Marine Engineers, 69, New York, N.Y., 1962 (reprinted by the International Nickel Co., Inc., N.Y.)

21. Schrieber, C.F., B.D. Oakes, A.L. Whitted, and F.H. Coley. Behavior of Metals in the Desalination Environment, 5th Progress Report. Materials Protection and Performance, 12 (8): 32, August 1973.

22. Gegner, P.J. and W.L. Wilson. Corrosion Resistance of Titanium and Zirconium in Chemical Plant Exposures. Corrosion 15 (7): 341t-350t, July 1959.

U S GOVERNMENT PRINTING OFFICE 1979 -261-167/129